

A Study on Thermal Characteristics of Epoxy Composites Filled with Natural Fiber and Particulate

A Thesis Submitted in Partial Fulfilment of the Requirement for the Degree of

Master of Technology
(By Research)

In

Mechanical Engineering

By

Mimosha Dash



**Department of Mechanical Engineering
National Institute of Technology
Rourkela(India)**

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Under the supervision of

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Associate Professor
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**Department of Mechanical Engineering
National Institute of Technology
Rourkela (India)**

2016



Dedicated to
My Beloved
Grand Father & Grand Mother



**National Institute of Technology
Rourkela**

C E R T I F I C A T E

This is to certify that the thesis entitled “*A Study on Thermal Characteristics of Epoxy Composites Filled with Natural Fiber and Particulate*”, submitted by **MIMOSHA DASH** to National Institute of Technology, Rourkela for the award of the degree of **Master of Technology**(By Research)in *Mechanical Engineering* is an authentic record of research work carried out by her under my guidance and supervision.

The work incorporated in this thesis has not been, to the best of my knowledge, submitted to any other University or Institute for the award of a degree or diploma.

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ABSTRACT

The research reported in this thesis broadly consists of three parts: The first part provides the description of the materials used and the details of the experiments that are carried out during this research. It also presents the test results in regard to the physical, mechanical and micro-structural characteristics of the epoxy filled with micro-sized teak wood dust and/or short banana fibers. The second part is about the development of a theoretical heat conduction model based on which a mathematical correlation has been proposed for estimation of effective thermal conductivity of polymer composites with uniformly distributed micro-sized particulate fillers. In this part, the correlation is validated for teak wood dust (TWD) filled polymers through numerical analysis and experimentation. The last part has presented the experimental results related to the effective thermal conductivity of hybrid composites filled with teak wood dust and short banana fibers (SBF). This part has also reported in regard to their other thermal characteristics such as glass transition temperature (T_g) and coefficient of thermal expansion (CTE).

The findings of this research suggest that by incorporation of micro-sized TWD particulates into epoxy resin, its effects, as expected are achieved in the form of modified physical, mechanical and thermal properties. Due to the presence of TWD micro-fillers, changes in their heat conduction behavior are seen. When TWD is added in epoxy matrix, the effective thermal conductivity of the composite is reduced as wood dust is insulative in nature. Additional reinforcement of short banana fibers in epoxy-TWD composite system further reduces the effective conductivity while enhancing the strength properties. Effects of TWD and SBF on the glass transition temperature and coefficient of thermal expansion of epoxy resin are also found to be substantial. With light weight, lowered thermal expansion coefficient and improved insulation capability, the teak wood dust and short banana fiber reinforced epoxy composites can be used for applications such as insulation boards, food containers, thermos flasks, refrigeration industry, building materials, interiors of air crafts and automobiles etc.

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CHAPTER 1

INTRODUCTION

Chapter 1

INTRODUCTION

1.0 Background and Motivation

With the growing awareness of global warming and fossil fuel depletion, agreements are being made to increase the production of sustainable energy and to improve the energy efficiency of engineering components and physical infrastructure so as to reduce greenhouse gas emissions worldwide. The potential impact of increasing the energy efficiency and thereby reducing the greenhouse gas emission through better thermal insulation of engineering structures and buildings is enormous. To achieve this, the most obvious approach would be to apply thicker layers of readily available thermal insulation materials. But this can locally result in the use of undesirable thick constructions and related changes in building methods, not to mention the loss of valuable floor space and the related higher costs. Instead of this option, novel types of affordable and processable thermal insulation materials with improved efficiencies are being considered [1]. Among the best thermal insulation materials available today are vacuum insulation panels [2] and aerogel based materials [3]. The low thermal conductivities of these materials originate from having a reduced gas phase conductivity based on vacuum and on pore sizes of below 100 nm. But in spite of their high thermal insulation efficiencies, these materials suffer from processability and pricing issues that restrict their wide spread utilization [3,4].

Requirement of Thermal Insulation

Insulation to retard the flow of heat is important for comfort and in some cases, for the survival of humans and animals. Insulation also prevents damage to various articles by freezing or by high temperatures, and lowers heating and cooling costs. Contrary to popular belief, insulation is not used to keep cold from

entering an area, but is used only to retard the flow of heat. Heat can flow in one direction only -- from a warmer body to a less warm body--so insulation serves to retard this flow. In a refrigerator, for example, insulation slows down the flow of heat from the room air to the interior of the refrigerator. In a building, insulation keeps heat in during winter and out during summer.

Air is a poor conductor of heat and when trapped in a hollow area is an excellent insulator. Other insulating materials, some of which depend on air pockets for much of their insulating effect, include mineral wool, fiberglass, asbestos, wood, concrete, vegetable fiber, vermiculite and foamed plastics such as polystyrene. These substances retard the conduction and convection of heat. Aluminum sheets and aluminum foils, on the other hand, prevent the radiation of heat, by reflecting it back towards its source.

Buildings can be insulated by using materials that themselves are good insulators or by leaving spaces in walls and ceilings and filling the spaces with an insulating material. Such simple building materials as the snow blocks of Eskimo igloos, the straw of thatched roofs and the sun-baked clay of houses in Northern Africa, the Middle East and Latin America provide good insulation. Many parts of our country too experience large changes in temperature from season to season. So, there is a great need for building materials with insulating properties with emergence of new technologies. The scenario in industries of all kinds, transportation, entertainment sector and even in the field of medical services is much the same.

1.1 Thermal Insulation

Heat flow is an inevitable consequence of contact between objects of differing temperature. Insulation provides a region of retarded heat flow in which thermal conduction is reduced or radiation is reflected rather than absorbed by the lower-temperature body. Thermal insulation thus is the reduction of heat transfer (the transfer of thermal energy between objects of

differing temperature) between objects in thermal contact or in range of radiative influence. This can be achieved with specially engineered methods or processes, as well as with suitable object shapes and materials.

Insulations are those materials or combinations of materials which retard the flow of heat energy by performing one or more of the following functions:

1. Conserve energy by reducing heat loss or gain.
2. Control surface temperatures for personnel protection and comfort.
3. Facilitate temperature control of process.
4. Prevent vapour flow and water condensation on cold surfaces.
5. Increase operating efficiency of heating/ventilating/cooling, process steam and power systems in commercial /industrial installations.
6. Prevent damage to equipment from fire or corrosive atmospheres.
7. Reduce emissions of pollutants to the atmosphere.

1.2 Types of Thermal Insulation

- Fibrous Insulation - composed of small diameter fibers which finely divide the air space. The fibers may be perpendicular or parallel to the surface being insulated and they may or may not be bonded together. Silica, rock wool, slag wool and alumina-silica fibers are used. The most widely used insulations of this type are glass fiber and mineral wool. Glass fiber and mineral wool products usually have their fibers bonded together with organic binders that supply the limited structural integrity of the products.
- Cellular Insulation - composed of small individual cells separated from each other. The cellular material may be glass or foamed plastic such as polystyrene (closed cell), poly-isocyanurate and elastomers.
- Granular Insulation - composed of small nodes which may contain voids or hollow spaces. It is not considered a true cellular material since gas can be transferred between the individual spaces. This type may be produced as a

loose or pourable material, or combined with a binder and fibers or undergo a chemical reaction to make a rigid insulation. Examples of these insulations are calcium silicate, expanded vermiculite, perlite, cellulose, diatomaceous earth and expanded polystyrene.

The conventional engineering materials are often unable to meet the requirement of these special properties like high strength, low density and low conductivity. The demand for structurally stable, cost effective and light-weight insulation materials is therefore increasing day by day. This is exactly why industrial and research efforts are directed towards the development of plastic foams and/or polymer matrix composites, eventually allowing the production of a new class of affordable, highly efficient thermal insulation materials. At present, the widely used insulation materials in the industries are based either on glass fibers, which are derived from the silica sources or on other synthetic fibers derived from the petroleum based resources. Unfortunately, glass fiber based materials are known to have carcinogenic effects [5] and petroleum based materials are non-renewable. This opens up another option i.e. polymer composites based on natural fibers.

Ironically, there is no widely accepted definition for a composite material. It can however be defined as any multiphase material that is artificially made and exhibits a significant proportion of the properties of the constituent phases. The constituent phases of a composite are usually of macro-sized portions, differ in form and chemical composition and essentially insoluble in each other. The properties of composites are a function of the properties of the constituent phases, their relative amounts and size-and-shape of the dispersed phase.

1. 3 Composite Materials

Composites are combinations of two or more materials in which one or more of the materials, called the reinforcing phase, are in the form of fibers or particles and are embedded in the base material called the matrix phase. The primary functions of the matrix are to transfer stresses between the reinforcing

fibers/particles and to protect them from mechanical and/or environmental damage whereas the presence of fibers/particles in a composite improves its mechanical properties such as strength, stiffness etc. A composite is therefore a synergistic combination of two or more constituents that differ in physical form and chemical composition and which are insoluble in each other. The objective is to take advantage of the superior properties of both materials without compromising on the weakness of either.

Composites have successfully substituted the traditional materials in several light weight and high strength applications. The reasons why composites are selected for such applications are mainly their high strength-to-weight ratio, high tensile strength at elevated temperatures, high creep resistance and toughness. Typically, in a composite, the reinforcing materials with low densities are strong while the matrix is usually a ductile or tough material. If the composite is designed and fabricated correctly it combines the strength of the reinforcement with the toughness of the matrix to achieve a combination of desirable properties not available in any single conventional material. The strength of the composites depends primarily on the amount, arrangement and type of fiber and/or particle reinforcement in the resin.

1.3.1 Types of Composites

Broadly, composites can be classified into three groups on the basis of matrix material. They are:

- a) Metal Matrix Composites (MMC)
- b) Ceramic Matrix Composites (CMC)
- c) Polymer Matrix Composites (PMC)

a) Metal Matrix Composites:

Metal matrix composites have many advantages over monolithic metals like higher specific modulus, higher specific strength, better properties at elevated temperatures and lower coefficient of thermal expansion. Because of these

attributes metal matrix composites are under consideration for wide range of applications viz. combustion chamber nozzle (in rocket, space shuttle), housings, tubing, cables, heat exchangers, structural members etc.

b) Ceramic Matrix Composites:

One of the main objectives in producing ceramic matrix composites is to increase the toughness. Naturally it is hoped and indeed often found that there is a concomitant improvement in strength and stiffness of ceramic matrix composites.

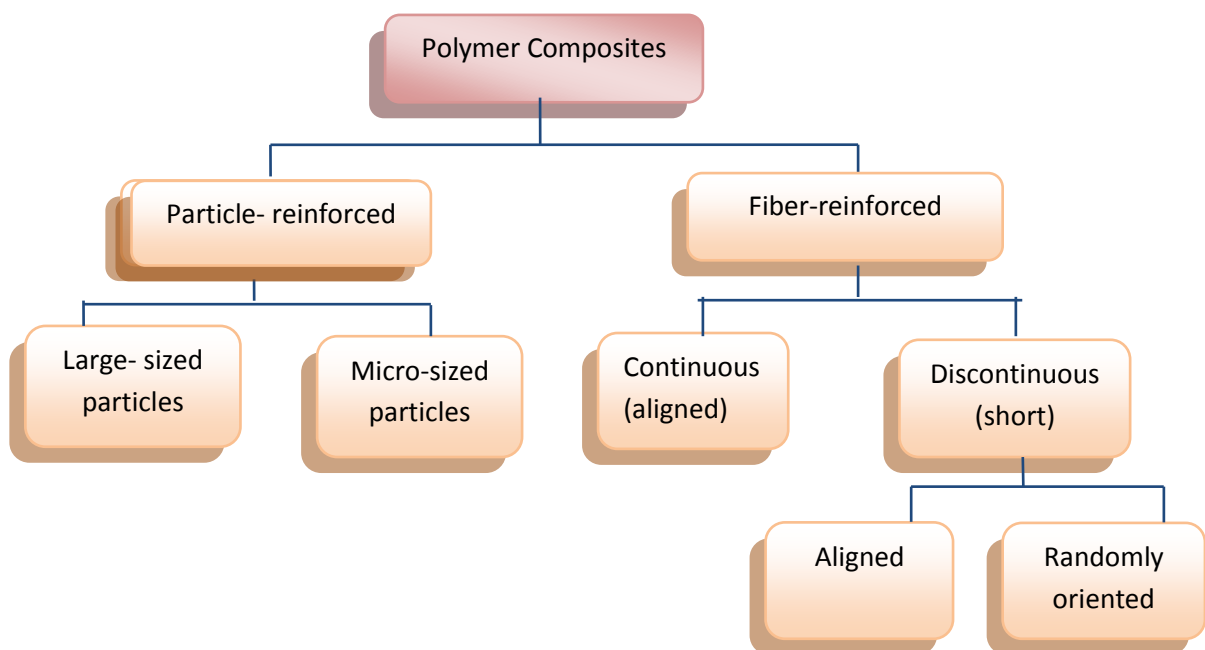


Figure 1.1 Classification of composites based on reinforcement type

c) Polymer Matrix Composites:

Most commonly used matrix materials are polymeric. The reasons for this are two-fold. In general the mechanical properties of polymers are inadequate for many structural purposes. In particular their strength and stiffness are low compared to metals and ceramics. These difficulties are overcome by reinforcing

other materials with polymers. Secondly, the processing of polymer matrix composites need not involve high pressure and does not require high temperature. Also equipment required for manufacturing polymer matrix composites are simpler. For this reason polymer composites developed rapidly and soon became popular for structural applications. Polymer composites are used because overall properties of the composites are superior to those of the individual polymers. They have a greater elastic modulus than the neat polymer but are not as brittle as ceramics.

Broadly, polymer composites can be classified into two groups on the basis of reinforcing material. They are:

- Fiber reinforced polymer (FRP)
- Particle reinforced polymer (PRP)

Fiber reinforced polymer

Common fiber reinforced composites are composed of fibers and a matrix. Fibers are the reinforcement and the main source of strength while matrix glues all the fibers together in shape and transfers stresses among the reinforcing fibers. The fibers carry the loads along their longitudinal directions. Sometimes, filler might be added to smooth the manufacturing process, impart special properties to the composites or to reduce the product cost. Common fiber reinforcing agents include asbestos, carbon/graphite fibers, beryllium, beryllium carbide, beryllium oxide, molybdenum, aluminium oxide, glass fibers, polyamide, natural fibers etc. Similarly common matrix materials include epoxy, phenolic resin, polyester, polyurethane, vinyl ester etc. Among these resin materials, polyester is most widely used. Epoxy, which has higher adhesion and less shrinkage than polyesters, comes in second for its high cost.

Particle reinforced polymer

Particles used for reinforcing include ceramics, small mineral particles, metal powders such as aluminium and amorphous materials including polymers and

carbon black. Particles are used to increase the modulus and to decrease the ductility of the matrix. Particles are also used to reduce the cost of the composites. Reinforcements and matrices can be common, inexpensive materials and are easily processed. Some of the useful properties of ceramics and glasses include high melting temperature, low density, high strength, stiffness, wear and corrosion resistance. Many ceramics are good electrical and thermal insulators. Some ceramics have special properties; some are magnetic materials; some are piezoelectric materials and a few special ceramics are even superconductors at very low temperatures. However, ceramics have one major drawback: they are brittle. An example of particle-reinforced composites is an automobile tyre, which has carbon black particles in a matrix of poly-isobutylene elastomeric polymer.

1.4 Introduction to the Research Topic

The present piece of research is basically an experimental investigation focused on the thermal characteristics of a new class of polymer composites filled with natural fibers and particulates. Effective thermal conductivity is an important parameter for characterization of thermal properties of particulate-filled polymer composite systems. Various complicated factors affect the thermal conductivity of such materials. In view of this, a theoretical heat transfer model for filled polymer based on the law of minimal thermal resistance and equal law of specific equivalent thermal conductivity is considered. A mathematical correlation based on this one dimensional conduction model for estimation of effective thermal conductivity of particulate filled polymer composites with different filler concentrations is proposed. The correlation is validated through numerical and experimental investigation for a set of polymer-particulate composites. Effect of additional reinforcement of short natural fibers on thermal conductivity of the polymer is studied. Further, the effects of fillers on other thermal characteristics of base polymer such as glass transition temperature and coefficient of thermal expansion are also studied.

1.5 Thesis Outline

The remainder of this thesis is organized as follows:

- Chapter 2:** Includes a literature review designed to provide a summary of the base of knowledge already available involving the issues of interest. It presents the research works of previous investigators on fiber and particulate reinforced polymer composites with emphasis on their thermal conduction behavior.
- Chapter 3:** Includes a description of the raw materials and the test procedures. It presents the details of fabrication and characterization of the composites under investigation.
- Chapter 4:** Presents the test results in regard to the physical, mechanical and micro-structural characteristics of the composites under study.
- Chapter 5:** Presents the development of a mathematical correlation for estimation of effective thermal conductivity of a class of particulate filled polymer composites. It also presents the results of analytical, numerical and experimental investigation on the thermal characteristics of the composites under study.
- Chapter 6:** Provides a summary of the findings of this research work; outlines specific conclusions drawn from both the experimental and analytical efforts and suggests ideas and directions for future research.

CHAPTER 2

Literature Review

Chapter 2

LITERATURE REVIEW

In the current chapter, the summary of the literature surveyed during this research has been presented. The purpose of this literature survey is to provide background information on the issues related to the present investigation and thereby to outline the objectives. This treatise thus embraces various aspects of fiber and particulate filled polymer composites with a special reference to their thermal characteristics such as conductivity, glass-transition temperature (T_g), coefficient of thermal expansion (CTE) etc. It includes reviews of available research reports on:

- Composite Materials for Thermal Insulation
- Particulate Filled Polymer Composites
- Polymer Composites with Natural Fibers
- Thermal Conductivity of Particulate Filled Polymers
- Thermal Conductivity of Fiber Reinforced Polymers
- Thermal Conductivity Models
- T_g and CTE of Polymer Composites

At the end of the chapter a summary of the literature survey and the knowledge gap in earlier investigations are presented. Subsequently the objectives of the present research work are outlined.

2.1 Composite Materials for Thermal Insulation

The building sector is moving towards new approaches to energy efficient design, which includes not only the decrease of the thermal transmittance of the building envelope but also the improvement and use of natural and locally available building materials. Materials based on natural fibers from renewable raw material resources are now becoming increasingly popular. Due to its low mass density and cell structure, they show very good sound and thermal insulation properties, often better and more

advantageous than synthetic fibers. A great advantage of the insulation based on natural fibers is not only the low value of thermal conductivity but also the natural character of input fibers. Another advantage is that it is a renewable material which does not place any significant strain on the environment. For example, when compared with mineral wool, the insulation based on natural fibers has comparable and sometimes even better thermal technical characteristics.

Thermal insulation plays an important role in contributing to the energy savings in the building by heat gains and losses through the building envelope [6]. Thermal insulation materials are results of research and developmental efforts devoted to make life more comfortable. Because the molecular density of gases is lower than that of bulk materials, heat transfer from one wall surface to the other is suppressed by the incorporation of a gas phase. Fibrous materials like glass fiber or rock wool are cost-effective and gas-foamed plastics such as urethane or phenolic foam exhibit good thermal stability in addition to thermal insulation [7,8]. The need for thermal insulation is not only very critical for technological development, but many industrial processes would not be possible without adequate thermal insulation. Thermal insulation helps to reduce power consumption during processing of materials and also protects the surrounding space from heat in the processing environment.

At present, bricks are used as the main wall materials in India, which causes a huge amount of farmland to be destroyed every year. Meanwhile, high energy consumption and serious environmental pollution for using bricks as the wall materials have great effect on the development of the building industry. To protect ecology and reduce energy consumption for buildings, a lot of attention is being paid to develop the environment-friendly and energy-efficient building materials. Because energy efficiency will be evaluated not only based upon heating demand, but also according to the primary energy demand, the ecological properties of the building materials for the whole assessment has become essential. The demand for green building materials is rising sharply, especially insulating materials from renewable resources [9]. It has been reported that typical thermal conductivity values for cellulose insulation materials are between 40 to 50 W/mK [10].

Many investigations have been carried out with natural materials and have shown that they are comparable with standard building materials. So far, many researchers have studied thermal insulation materials from ligno-cellulosic fibers. Khedari et al. [11,12] developed a new low cost particle-board from durian peel and coconut coir mixture with a lower thermal conductivity, which was effective for energy saving when used as ceiling and wall insulating material. Xu et al. [13] presented a low-density binderless particle-board from kenaf core using steam-injection pressing, with a thermal conductivity similar to those of insulation material (i.e. rock wool).

An environment-friendly thermal insulation material binder-less cotton stalk fiberboard was developed from cotton stalk fibers without resins and other chemical additives and exposed that it can successfully compete with other insulation materials [14]. The research article of Agoudjil et al. [15] shows that the date palm wood is a good candidate for the development of efficient and safe insulating materials when compared to other natural materials. The low thermal conductivity and fibrous character of the majority of organic materials contribute to a significant improvement in the insulation properties after incorporation in the structure of the exterior building envelope. A study has been reported on the development of insulation materials from sheep wool fibers with comparable properties as that of conventional materials [16]. In another recent study, authors highlighted the quantity issues of alternative sheep wool materials available in the market to meet the demand for the building sector, although sheep wool materials are very good insulators [17]. The use of industrial crops and food crop by-products as raw materials for composites to be used in building applications is an interesting alternative to conventional products [18-21]. Although much of the research has been focused on the reinforcement in composites and the development of high and medium density boards, there is an increasing interest in their use in natural thermal insulations [9].

Evon et al. [22] manufactured a new class of thermal insulation fiberboards by thermo-pressing from sunflower mixed with proteins and ligno-cellulosic fibers, acting respectively as binder and reinforcing fillers in what could be considered as a natural composite. The result showed that the heat insulation properties improved with

decreasing fiberboard density and thermal conductivity of the least dense fiberboard was only 0.0885 W/mK at 25⁰C. Mati-Baouche et al. [23] carried out a research to develop a new insulating bio-based composite made with chitosan and sun-flower's stalk particles. Panyakaew and Fotios [24] described the production of low density thermal insulation boards made from coconut husk and bagasse without the use of chemical binding additives. Both insulation boards have thermal conductivity values ranging from 0.046 to 0.068 W/mK which were close to those of conventional insulation materials such as cellulose fibers and mineral wool. More similar bio-based insulation materials have been developed from agricultural straw and natural adhesives [25,26,13]. Binici et al. [27] had utilized sunflower stalks and cotton textile waste to manufacture thermal insulation materials with epoxy as binder. An experimental work concluded that samples having sunflower stalk with cottony tissue had low heat transfer coefficients. Korjenic et al. [9] developed a new organic thermo insulating material from renewable resources (jute, flax and hemp) and binders with comparable building physics and mechanical properties to conventional insulations materials. Other authors [28-30] have been studying the technical potential of using other types of synthetic resins for producing bio-based thermal insulation materials, such as soy protein-based adhesive, urea-formaldehyde adhesive and hybrid organic–inorganic binder. Xiaoyan Zhou et al. [31] reported that the use of straws in building will not only solve the problem of straw as a waste, but also will help in building houses having significantly low impact on the environment without sacrificing most of comforts. Goodhew and Griffiths [32] showed that the straw-bales offer good insulation values of 0.067 W/mK, which is much lower than that of wood and other building raw materials. There are undoubtedly strong reasons to believe that developed alternative products from agricultural straws are promising in a thermal insulating perspective [18].

2.2 On particulate filled polymer composites

The technique to improve mechanical performance of materials by dispersing particles in a matrix has been applied to ceramic, metal and polymer matrices and these

materials are called particulate-reinforced composites. Polymers filled with such particulate fillers are being widely acceptable these days in several industries. Incorporating organic and/or inorganic fillers into a matrix enhances various physical properties of the materials such as mechanical strength, elastic modulus and heat transfer coefficient [33,34]. In general, the mechanical properties of particulate filled polymer composites depend strongly on size, shape and distribution of filler particles in the polymer matrix [35]. The addition of particles into thermoset polymer has been demonstrated to provide an effective enhancement of the strength and toughness of composite materials [36]. Silica, alumina and glass particles have been used as filler and epoxy–silica based composites are widely used in electronics, automotive and aerospace industries to dissipate heat and prevent electrostatic accumulation [37]. The interaction of the fillers with the composite matrix is particularly important when irreversible or energy dissipation processes are active, as in the case of plastic deformation and fracture. The particle addition tends in general to increase the stiffness of the matrix, leading to the enhancement of Young's modulus [38,39-42], toughness [38,41,42], tensile [39] and flexural strength [43]. Other properties that exhibit a remarkable improvement when using particles as third phase are the compressive strength [44,45], impact strength [39,46] and glass transition temperature [43], in addition to improve the fiber–matrix interface conditions [46] especially at high temperatures [38]. Particle size and weight fraction for matrix reinforcement are selected judiciously to provide specific properties for the composite [47].

The fillers are essentially added to the polymers to improve the cost reduction, improved processing, density control, optical effects, control of thermal expansion and thermal conductivity, electrical properties, magnetic properties, flame retardancy and improved mechanical properties such as hardness and wear resistance. For example, in cable applications, fillers such as meta-kaolinite are used to provide better electrical stability while others like alumina trihydrate are used as fire retardants. Each filler type has different properties and these in turn are influenced by the particle size, shape and surface chemistry. Moreover, most of the particulate fillers are inorganic and

polar, which may give rise to poor compatibility with hydrocarbon polymers and processing problems among various other effects [48].

Hard particulate fillers consisting of ceramic or metal particles and fiber fillers made of glass are being used these days to dramatically improve the wear resistance even up to three orders of magnitude [49]. Various kinds of polymers and polymer matrix composites reinforced with metal particles have a wide range of industrial applications such as heaters, electrodes [50], composites with thermal durability at high temperatures [51] etc. These engineering composites are desired due to their low density, high corrosion resistance, ease of fabrication and low cost [52,53,35]. Similarly, ceramic filled polymer composites have been the subject of extensive research in last two decades. Along with fiber-reinforced composites, the composites made with particulate fillers have been found to perform well in many real operational conditions.

When silica particles are added into a polymer matrix to form a composite, they play an important role in improving electrical, mechanical and thermal properties of the composites [54,55]. Recently, reduction in particle size is being adopted by various researchers and many studies have focused on how single particle size affects mechanical properties of the composites [56-62]. Yamamoto et al. [63] reported that the structure and shape of silica particles have significant effects on the mechanical properties such as tensile strength, fracture and fatigue resistance. Nakamura et al. [64-66] discussed the effects of size and shape of silica particles on the strength and fracture toughness based on particle-matrix adhesion. Moloney et al. [67-69] and Adachi et al. [70] found that the mechanical properties of epoxy composites were dependent on volume fraction of particles. Furthermore, effects of different particles of micron size magnitude and nanoparticles on the properties of the composites were discussed by Yuan et al. [71] and Ng et al. [72].

It has also been found that composites with alkali treated fillers (wood particles) show higher tensile strength as suggested by Islam et al. [73]. The increase of tensile properties is possibly due to the fact that the alkali treatment improves the filler

surface adhesion characteristics and partly removes the relatively weak lignin portion of the wood particle. In other words alkali treatment reduces filler size and thereby increases the aspect ratio leading to better filler-matrix interface adhesion and development in mechanical properties. So, treated wood dust composites provided higher tensile strength compared to that of composite reinforced with untreated wood particle. The tensile strength of this type of composite with chemically treated wood dust has been investigated and similar results have been found by other investigators [74-76]. The addition of wheat flour and sawdust to PP clearly altered its mechanical, morphological, physical and thermal properties considerably. The wheat flour composites exhibited better mechanical properties than the wood sawdust composites due to their smaller particle size that provided higher surface area for efficient filler-matrix interfacial adhesion [77]. Maiti and Singh [78] examined influence of wood flour size that was compounded with HDPE followed by extrusion moulding.

In 2008, Fu et al [79] investigated in detail the effect of particle size, particle/matrix interface adhesion and particle loading on the mechanical properties of particulate filled polymer composites. Tajvidi et al. [80] studied extruded composite made from Reed flour-PP with particle sizes ranging from 20-40 mesh to smaller than 100 mesh sizes. Yang et al. [81] prepared a composite sample with PP as the matrix and rice husk flour as the reinforcing filler and studied the physical, mechanical and morphological properties. Inch-Long Ngo et al. [82] investigated the effect of filler shape on the effective thermal conductivity.

The incorporation of mineral particles into polymers can improve their mechanical properties. For example, the high stiffness of mineral particles can lead to increase in composite's stiffness [83]. Micron-sized particles usually cause a reduction in impact resistance [84]. Decreasing the interfacial strength could cause the interfacial debonding of particles from the matrix [85] and, as a consequence, the tensile strength of the composite could be reduced. Interfacial debonding can also cause shear yielding of the matrix around the particles [86]. Lin et al. [87] demonstrated that the debonding of particles from the polymer matrix can increase the impact energy, even though the tensile strength declined. Conversely, when particles are bonded perfectly to the

polymer, this can cause the matrix to craze around the particles such that, as a consequence, impact resistance is reduced [88-91]. Liu and Sun [92,93] and Jiang et al. [94] developed the micro-mechanics based models of particle reinforced composites by taking account of the interphase between particles and matrix and discussed the particle size effect on deformation due to interphase.

Many studies have been conducted on the mechanical properties of particulate-filled polymer composites. It has reported that stiffness can be readily improved by adding either micro or nanoparticles since rigid inorganic particles generally have a much higher stiffness than polymer matrices [95-103]. However, strength strongly depends on the stress transfer between the particles and the matrix. For well-bonded particles, the applied stress can be effectively transferred to the particles from the matrix [104]; this clearly improves the strength [95,105-108]. However, for poorly bonded micro-particles, strength reduction occurs by adding particles [96-98,109-115].

Polymer composites are noted to show mechanical properties which depend on time, rate and temperature [116]. Viscoelastic moduli are mainly governed by the volume fraction of particles [117] and strain rate has important effects on matrix/particulate interface adhesion and other mechanical properties [118-121]. The strength of micro-particle-filled composites decreases with particle content. The reverse is also true, i.e. in micro-size, strength increases with particle content. This contradiction is because, besides particle size and loading, the particle/matrix interfacial adhesion also significantly affects the strength of particulate composites [122]. The incorporation of a small amount of BaSO₄ particles in PP does not significantly affect the yield strength of the composites [88], whereas a relatively high content of BaSO₄ particles (>16 wt%) has a negative effect on the yield strength. It is noted that the yield strength depends on the interfacial modification.

2.3 Polymer Composites with Natural Fibers

In recent years, the interest of scientists and engineers has turned over on utilizing plant fibers as effectively and economically as possible to produce good quality fiber-reinforced polymer composites for structural, building and other needs. It is because

of easy availability of natural fibers that has led to the development of alternative materials instead of conventional or man-made ones. Many types of natural fibers have been investigated for their use in polymer such as wood fiber [123], sisal [124], pineapple [125], jute [126] and banana [127].

Bax and Mussing [128] investigated the mechanical properties of PLA reinforced with cordenka rayon fibers. Waikamboo and Ansell [129] evaluated the physical and mechanical properties of the natural fiber composites to assess their serviceability. It is remarkable to note that natural fibers such as jute, coir, banana, sisal etc. are richly available in developing countries like India, Sri Lanka and some of the African countries, but are not optimally utilized. At present, these fibers are used for the production of yarns, ropes, mats and matting as well as in making fancy articles like wall hanging, table mats, handbags and purses. Fibers such as cotton, banana and pineapple are also used in making cloth in addition to being used in the paper industry. It is a well-known fact that banana is one of the oldest cultivated plants in the world. Banana-trees produce generally 30 large leaves (almost 2m long and 30-60cm wide) [130]. The cross sectional area of the banana fiber was investigated by using optical laser beam equipment by Murali et al [131], and it was found to be 0.3596mm^2 . A truck model 'Manaca' was developed and tested by Al-Qureshi [132], using banana fiber and epoxy resin. However, some special and critical panels were made of hybrid composites of glass/banana fibers. The vehicle went through many years of road performance tests and provided excellent results.

Banana fiber has already been used as reinforcement with various thermoplastics, namely PP, Low Density Polyethylene (LDPE), High Density Polyethylene (HDPE), Polystyrene (PS) and Poly Vinyl Chloride (PVC). The influence of sisal, banana, jute and flax fiber morphology on the mechanical properties was studied by Kristina et al [133]. The composites are produced by extrusion method. Of the various fibers compared, sisal fiber has better impact properties, because it has better elongation to break. The performance evaluation of the hybrid composite of banana/glass fiber with PP was carried out by Sushanta et al [134]. The bio-degradability of banana, pineapple and bamboo fibers reinforced in PP matrix was studied by Sanjay et al [135].

A comparative study of fibers from cotton, rice straw, bagasse and banana plant as reinforcement in LDPE was conducted by Youssef et al [136]. The study revealed that better tensile and thermal properties are achieved when maleated LDPE is used as a matrix, when compared to untreated LDPE. Further, it also shows that the chemical composition of the natural fibers has a strong influence on the mechanical properties of the composite. The influence of the fiber content, fiber loading and hybrid effect on the mechanical properties of banana/glass hybrid composite was studied by Anshida et al [137]. They reported that the modification of the banana fiber by alkali, benzylation and PSMA treatments improved the interface adhesion and hence, the mechanical properties of the composite.

Composite of various thermoset matrices (Polyester, Phenol formaldehyde, Urea Formaldehyde and Epoxy) reinforced with banana fibers were investigated by various researchers in the past [138-142]. Banana fiber reinforced polyester matrix was extensively investigated by Laly et al [138]. The study shows the effect of the fiber length and content on mechanical properties of the composite. The investigation shows that the fiber length of 30-40 mm and 40 % volume content has better mechanical properties. Another study by Sreekumar et. al. [139] also showed that the maximum diffusion, sorption and permeability coefficient are achieved at 50vol%. The effect of chemical treatment on the flexural, impact and water absorption properties of woven banana-polyester composite was analyzed by Jannah et al. [140]. The effect of the layering arrangement on the storage modulus, loss modulus and damping property of banana/sisal hybrid composite as the function of temperature and frequency was studied by Mariers et al. [141]. The comparative study of Phenol Formaldehyde (PF) reinforced with banana and glass fibers showed that optimum mechanical properties are achieved at different fiber lengths. The interface adhesion was better between banana fiber and PF when compared with glass fiber and PF, which was determined from the single fiber pull-out test. It also revealed that the specific properties of the banana fiber- PF are superior to those of the glass fiber- PF composite [142].

2.4 Thermal Conductivity of Particulate Filled Polymer Composites

In 1960s and 1970s, considerable work has been reported on the subject of heat transport in polymers by Hennig and Knappe [143], Hansen and Ho [144], Peng and Landel [145], Choy and Young [146] etc. Later on, Tavman [147] successfully provided a criterion for anisotropic heat conduction behavior of polymers by changing their molecular orientation. In a recent work, Griesinger et al. [148] have reported that the thermal conductivity of polyethylene increases from 0.35 to 50 W/m-K by keeping an orientation ratio of 50. However, most of these studies were confined to the thermal behavior of neat polymers and not to their composites.

Since it is not always possible to fabricate a composite keeping the molecular orientation as per choice, a more practical method to increase the thermal conductivity of the polymer is by adding thermally conductive particles or fibers onto it. Lot of work has been reported to improve the thermal conductivity of polymers by incorporating conductive fillers. Most of them include the experimental determination of effective thermal conductivity of particulate filled polymer composites [149-153]. Metals are known for their high thermal conductivity, so they are being widely used as fillers in polymer composites. Sofian et al. [149] studied the effect of various metal powders like copper, zinc, iron and bronze on the thermal properties like conductivity, diffusivity and specific heat of HDPE matrix. Mamunya et al. [150] later reported the improvement in thermal conductivity of two different categories of polymers i.e. thermoplastic (polyvinyl chloride) and thermoset (epoxy) filled with copper and nickel particles. Though in 1990s, Tecke et al. [151] and Tavman [35] had already used copper powder as filler and measured the thermal conductivity of the composites by hot disc method, more recently copper has again been used by Luyt et al. [152] in LDPE. They have reported around 150% increase in the value of thermal conductivity for the composites filled with 24 vol% of copper. Subsequently, Tavman [153] took aluminium powders as filler and studied the thermal property of HDPE whereas later Boudenne et al. [154] gave an overview on the thermal conductivity of PP/aluminium composites. Silver too has a high potential to be used as filler because of its high thermal conductivity. The effect of silver particulates in epoxy was studied by

Bjorneklett et al. [155]. The filling of a polymer with metallic particles though resulted in increased thermal conductivity; simultaneous increase in density of the composites was also recorded, thus restricting the use of metal powders for light-weight applications.

Carbon-based fillers with high thermal conductivity and low density appear to be the most promising fillers. Graphite, carbon fiber and carbon black are well-known carbon-based fillers. Graphite is considered as the best conductive filler because of its good thermal conductivity and low cost [156]. Graphite with single graphene sheet shows intrinsically high thermal conductivity of about 800W/mK [157]. Expanded graphite, an exfoliated form of graphite with layers of 20-100nm thickness, has also been used in polymer composites [158]. Carbon fiber, typically vapor grown carbon fiber (VGCF), is another important carbon-based filler [159]. Studies conducted on modified thermal conductivity of polymer composites filled with carbon nanotubes have recently been reviewed by Han and Fina [160].

Metallic and carbon-based fillers are highly conductive as far as heat flow is concerned, but they are electrically conductive as well. There are certain areas where high thermal conductivity is required but at the same time electrical resistivity is of prime importance, like in electronic devices. In that case a balance is required that maximizes the thermal dissipation effect of the electrically insulating molding compound as well as which prevents the leakage of current across the conductors due to very low resistivity. Ceramic powder reinforced polymer materials have been used extensively for such applications because of their high thermal and low electrical conductivity. Some promising ceramic fillers such as SiC [161], Si₃N₄ [162], Sr₂Ce₂Ti₅O₁₆ [163], CeO₂ [164] AlN [165,166], Al₂O₃ [167,168] and ZnO [169,170] are in use to improve thermal conductivity of various polymers. It is observed from the above literature on ceramic particle filled polymers that there is an appreciable increase in thermal conductivity of the composite with increase in filler concentration whereas no significant changes are observed in electrical conductivity of such

composites. Incorporation of multiple fillers into the polymer matrix for the improvement of thermal conductivity has also been reported [171-173].

Factors other than the intrinsic thermal conductivity of the fillers such as shape, size, distribution and interconnectivity between the particles also decide the composite thermal conductivity. Boudenne et al. [154] studied the effect of two different particle sizes of aluminium filler and found that the composite filled with larger particle size shows significantly high thermal conductivity. It is due to the formation of more stable thermally conductive pathways in the matrix material. Similar behaviour was observed by Zhou et al. [174]. On the contrary, some authors have highlighted the higher heat transport ability of the composites filled with smaller particles [149, 175]. Boudenne et al. [176] too experimented with copper powders instead of aluminum and found better thermal conductivity with particles of smaller size. Weidenfeller et al. [177] studied the effect of the interconnectivity of the filler particles and its important role in improving the thermal conductivity of the composites. Tekce et al. [151] noticed the strong influence of the shape factor of fillers on thermal conductivity of the composite. Effects of interfacial thermal barrier resistance together with particle shape and size on the thermal conductivity was shown by Jiajun et al. [178].

Most of the research works to predict the thermal conductivity of polymer composites is on experimental basis, though some numerical studies have also been done to predict the effective thermal conductivity [179,180]. A numerical approach to evaluate the effective thermal conductivity of granular or fibrous reinforced composite materials was proposed by Veyret et al. [181], whereas Kumlutas and Tavman [182] have developed a numerical model for particulate filled polymers which shows good agreement with the experimental values.

2.5 Thermal Conductivity of Fiber Reinforced Polymer Composites

A great deal of research work has been reported by various authors on the thermal characteristics of fiber reinforced composites. Kim et al [183] studied the glass fiber reinforced poly-metal-phosphate matrix composites displaying excellent thermal insulation and mechanical properties. Schuster et al [184] investigated the effect of

three-dimensional fiber reinforcement on the out-of-plane thermal conductivity of composite materials. Using finite element models to better understand the behavior of the composite material, improvements to an existing analytical model were performed to predict the effective thermal conductivity as a function of the composite material properties and in-contact thermal properties. Zhidong et al [185] studied the dependence of thermal conductivity of nanotubes on the atomic structure, tube size, morphology and the defect. The roles of particle/polymer and particle/particle interfaces on the thermal conductivity of polymer/CNT nano composites are discussed in detail. Yüksel et al [186] studied the temperature dependence of effective thermal conductivity (ETC) for samples of binary, ternary and quadruple glass wools reinforced with aluminium foil. Hang et al [187] investigated thermal transport mechanisms and characterized when highly conductive fibers are embedded across the thickness of a three-dimensional polymer composite.

2.6 Thermal Conductivity Models

Several theoretical models and correlations have been established in the past by various researchers to predict the ETC of two phase mixtures and composites. The Maxwell-Garnett model or M-G model (Maxwell, 1873) is the first theoretical model used to describe the effective thermal conductivity of two component mixtures. Comprehensive review articles have discussed the applicability of many of these models [188,189]. For a two-component composite, the simplest alternatives would be with the materials arranged in either parallel or series manner with respect to heat flow, which gives the upper or lower bounds of effective thermal conductivity.

For the parallel conduction model:

$$k_c = (1 - \phi)k_m + \phi k_f \quad (2.1)$$

Where k_c , k_m , k_f are the thermal conductivities of the composite, the matrix and the filler respectively and ϕ is the volume fraction of filler.

For the series conduction model:

$$\frac{1}{k_c} = \frac{(1 - \phi)}{k_m} + \frac{\phi}{k_f} \quad (2.2)$$

The above two correlations (2.1, 2.2) are derived on the basis of the Rule of Mixture (ROM). Tsao [190] derived an equation relating the thermal conductivity of a two phase solid mixture to the conductivity of the individual components and two parameters which describe the spatial distribution of the two phases. By assuming a parabolic distribution of the discontinuous phase in the continuous phase, Cheng and Vachon [191] obtained a solution to Tsao's model that did not require knowledge of additional parameters. Agari and Uno [192] proposed a new model for polymers which takes into account parallel and series conduction mechanisms. According to this model, the expression that governs the thermal conductivity of the composite is:

$$\log k_c = \phi C_2 \log k_f + (1 - \phi) \log(C_1 k_m) \quad (2.3)$$

Where, C_1 , C_2 are experimentally determined constants of order unity. C_1 is a measure of the effect of the particles on the secondary structure of polymer, like crystallinity and the crystal size of the polymer. C_2 is the conductive chain formation potentiality of the particles. Bruggeman [193] derived an equation employing different assumptions for permeability and field strength for dilute suspension of spheres for a homogeneous medium and the implicit equation is given as:

$$1 - \phi = \left[\frac{k_c - k_f}{k_m - k_f} \right] \left(\frac{k_m}{k_c} \right)^{1/3} \quad (2.4)$$

Kanari model [194] which is a revised Bruggmen's equation is another empirical model using inorganic particles as filler. It presents a relationship between the thermal conductivity of composites and the volume fractions of filler .

$$1 - V_f = \frac{k_c - k_f}{k_m - k_f} \left(\frac{k_m}{k_c} \right)^{\frac{1}{(1+x)}} \quad (2.5)$$

Where V_f is the volume fraction of filler, k_c is the thermal conductivity of composite; k_f is the thermal conductivity of the filler; k_m is the thermal conductivity of the matrix; and x is the constant determined by sphericity of the filler and k_f/k_m .

Maxwell's equation [195] is an exact solution for the effective conductivity of randomly distributed and non-interacting spheres in a continuous medium. However, it does not take into account the mutual interaction of the particles thus, it is not a satisfactory treatment for composites in high volume fraction range.

$$k_c = k_m \left[\frac{k_f + 2k_m + 2\phi(k_f - k_m)}{k_f + 2k_m - 2\phi(k_f - k_m)} \right] \quad (2.6)$$

Where, k_c , k_m and k_f are thermal conductivities of composite, continuous-phase (matrix), and dispersed-phase (filler) respectively and ϕ is the volume fraction of the dispersed-phase. Lewis and Nielsen [196] derived a semi-theoretical model by modification of the Halpin-Tsai equation for a two phase system which assumes an isotropic particulate reinforcement and also takes into consideration the shape of particle as well as its orientation.

$$k_c = k_m \left[\frac{1 + AB\phi}{1 - B\phi\psi} \right] \quad (2.7)$$

$$\text{Where } B = \left[\frac{(k_f/k_m) - 1}{(k_f/k_m) + A} \right] \text{ and } \psi = 1 + \left[\frac{1 - \phi_m}{\phi_m^2} \right]$$

where, k_f is thermal conductivity of filler material and ϕ is the volume fraction of filler material. The value of A and ϕ_m for different shapes are provided in the Table 2.1 and 2.2 respectively.

Table 2.1 Value of A for various systems [196]

Type of dispersed phase	Direction of heat flow	A
Cubes	Any	2
Spheres	Any	1.5
Aggregates of spheres	Any	$(2.5/\phi_m) - 1$
Randomly oriented rods Aspect ratio=2	Any	1.58
Randomly oriented rods Aspect ratio=4	Any	2.08
Randomly oriented rods Aspect ratio=6	Any	2.8
Randomly oriented rods Aspect ratio=10	Any	4.93
Randomly oriented rods Aspect ratio=15	Any	8.38
Uniaxially oriented fibers	Parallel to fibers	2L/D
Uniaxially oriented fibers	Perpendicular to fibers	0.5

Table 2.2 Value of ϕ_m for various systems [196]

Shape of particle	Type of packing	ϕ_m
Spheres	Hexagonal close	0.7405
Spheres	Face centred cubic	0.7405
Spheres	Body centred cubic	0.60
Spheres	Simple cubic	0.524
Spheres	Random close	0.637
Rods and fibres	Uniaxial hexagonal close	0.907
Rods and fibres	Uniaxial simple cubic	0.785
Rods and fibres	Uniaxial random	0.82
Rods and fibres	Three dimensional random	0.52

2.7 T_g and CTE of Polymer Composites

Apart from heat dissipation problem, high coefficient of thermal expansion (CTE) of matrix material is also a major source of thermal failure in many applications where material is subjected to periodic heating and cooling due to thermal cycling. Iyer et al. [197] have recently reported significant reduction in CTE as the content of boron nitride is increased in polymer composites. Dey et al. [198] have studied the dependence of CTE on volume fraction of filler at ambient temperature. Yasmin et al. [199] have reported that, as the graphite concentration in epoxy increases to about 2.5 wt %, glass transition temperature (T_g) increases and CTE of the composite decreases, however on further addition of graphite i.e. 5 wt.%, the CTE of the composite starts increasing.

T_g and CTE play an important role in characterizing the thermal properties of polymer composites. Few works has been reported to resolve the problem of high CTE of polymers. Benito et al.[200] reported on CTE of TiO_2 filled EVA based nanocomposites and the influence of filler size on composites. Thomas et al. [201] studied the effect of $\text{Sm}_2\text{Si}_2\text{O}_7$ particles as reinforcement in two different matrix materials (polyethylene and polystyrene) on CTE. Li Sun et.al.[209] reported addition of small amount of Al_2O_3 into the $\text{WO}_3 + \text{ZrO}_2$ reaction system which was found to effectively increase the final densities and Young's moduli of the sintered composites along with a remarkable change in CTE of these composites. Devendra et.al. [202]

reported that with increase in the addition of filler materials to polymers, the thermal expansion coefficient of composites reduced. Dey et al. [203] investigated the thermal properties of silicon powder filled HDPE composites and reported that CTE of the composites showed strong dependence on silicon loading. Silica is used as the filler to reduce the CTE of under fill so as to match the CTE of the solder material to achieve high reliability [204]. Goyel et. al. [205] developed high performance polymer/AlN composites for electronic substrate applications and found that it has unique combination of anisotropic linear coefficient of thermal expansion and dielectric properties. Recently, Agrawal et.al. [206] have also reported on modified T_g and CTE of epoxy composites filled with AlN and Al_2O_3 . Similar findings have also been reported by Mishra et.al. [48] and Sahu [207] for epoxy based composites filled with solid glass microspheres and TiO_2 respectively.

2.8 Knowledge Gap in Earlier Investigations

In spite of a number of research works reported in the past, there is a huge knowledge gap that demands a well-planned and systematic research in this area of particulate filled polymer composites. An exhaustive review of the published literature reveals that:

- Most of the investigations are aimed at enhancing the thermal conductivity of the polymer by adding conductive fillers rather than attempting to improve its insulation capability.
- Most of the studies are for single filler composites and only very few papers have reported on the synergistic effects of two different kinds of fillers on the thermal properties.
- Reports are available in the literature on studies carried out on thermal behavior of polymer composites with metal and/or ceramic fillers but only very few reports are available on composites filled with natural fibers and particulates.

- Most of the works reported on thermal conductivity of particulate filled polymers are experimental in nature and reports available on numerical and analytical models are few.
- Though a few papers have been reported on thermal behavior of wood dust filled polymer composites, a possibility that the incorporation of both wood dust and short natural fibers in epoxy could provide a synergism in terms of improved insulation and modified mechanical properties has not been addressed so far.

In view of the above, the present work is undertaken to investigate on the thermal characteristics such as effective thermal conductivity, glass transition temperature, coefficient of thermal expansion etc. of epoxy matrix composites filled with micro-sized teak wood dust (TWD) and/or short banana fibers (SBF).

2.9 Objectives of the Present Work

The objectives of this work are outlined as follows:

- Development of theoretical models for estimation of effective thermal conductivity of polymer composites with micro-sized spherical fillers.
- Estimation of effective thermal conductivity of such particulate filled polymer composites using finite element method (FEM).
- Fabrication of different sets of epoxy composites filled with TWD and measurement of their thermal conductivities.
- Validation of the proposed theoretical model and FEM results by comparing the theoretical values with the measured values of effective thermal conductivity of the composites.
- Physical, mechanical and micro-structural characterization of the composites.
- Study on the effects of additional short banana fiber reinforcement on effective thermal conductivity of epoxy-TWD composites.

- Study on the effects of TWD on properties like glass transition temperature and coefficient of thermal expansion of these composites and exploring the possibility of their use as domestic/industrial insulation materials.

Chapter Summary

This chapter has provided

- An exhaustive review of research works on various aspects of particulate/fiber filled polymers reported by previous investigators
- The knowledge gap in earlier investigations
- The objectives of the present work

The next chapter describes the materials and methods used for the processing and characterization of the composites under this study.

CHAPTER 3

Materials and Methods

Chapter 3

MATERIALS AND METHODS

This chapter describes the materials and methods used for processing and characterizing the composites under this investigation. It presents the details of the tests related to the physical, mechanical, micro-structural and thermal characterization of the epoxy composites prepared for the present research.

3.1 MATERIALS

3.1.1 *Matrix Material*

Recently, polymer based composites are becoming more attractive due to their outstanding properties such as corrosion resistance, low coefficient of thermal expansion, wear resistance, thermal conductivity etc. Polymers are of particular interest due to their electrical resistivity, lightweight, ease of manufacturing and low cost [208]. Polymers are broadly classified into two types, namely thermoplastics and thermosets [209]. Each of them has its own individual chemical characteristics based on its molecular structure.

Thermoset matrices are formed due to an irreversible chemical transformation of the resin into an amorphous cross-linked polymer matrix. Due to huge molecular structures, thermoset resins provide good electrical and thermal insulation. They have low viscosity, which allows proper fiber wet out, excellent thermal stability and better creep resistance [210]. The most commonly used thermosets are epoxy, polyester, vinyl ester and phenolic resin. Thermoplastic polymers are associated through chain due to intermolecular forces and these forces permits thermoplastics to be remolded because the intermolecular interactions increase upon cooling and restore the bulk properties. These polymers are normally produced in one step and then are made into products in a subsequent process. Thermoplastic polymers can also be readily recycled

because each time they are reheated; they can again be reshaped or formed into a new article. Various types of thermoplastic polymers available in the market are acrylic, nylon, polyethylene, polypropylene, polystyrene, polyvinyl chloride, teflon etc.

Among the wide variety of thermosetting polymers available in the market, epoxy resins are widely being used for many advanced composites due to their excellent adhesion to a wide variety of fibers, superior mechanical and electrical properties and good performance at elevated temperatures. In addition, they have low shrinkage upon curing and good chemical resistance. Due to several advantages over other polymers as mentioned above, epoxy (LY 556) is chosen as the matrix material for the present research work. It chemically belongs to the ‘epoxide’ family and its common name is Bisphenol-A-Diglycidyl-Ether (commonly abbreviated to DGEBA or BADGE). Its molecular chain structure is shown in Figure 3.1. It provides a solvent free room temperature curing system when it is combined with the hardener tri-ethylene-tetra-amine (TETA) which is an aliphatic primary amine with commercial designation HY 951 (Figure 3.2). The LY 556 epoxy resin (Figure 3.3) and the corresponding hardener HY-951 are procured from Ciba Geigy India Ltd. for the present work. Table 3.1 provides some of the important properties of epoxy.

Table 3.1 Properties of epoxy resin [207]

Characteristic Property	Inferences
Density	1.1 gm/cc
Compressive strength	90 MPa
Tensile strength	58 MPa
Micro-hardness	0.085 GPa
Thermal conductivity	0.363 W/m-K
Glass transition temperature	98°C
Coefficient of Thermal expansion	62.83 ppm /°C
Electrical conductivity	0.105×10^{-16} S/cm

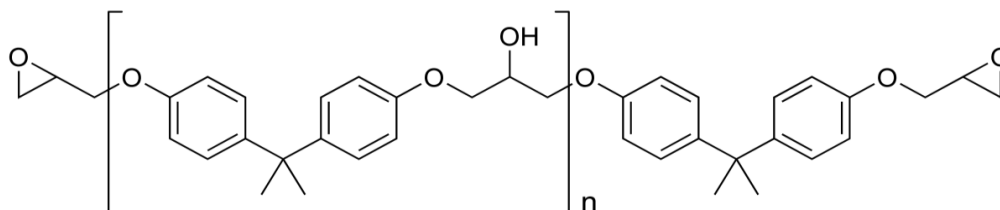


Fig. 3.1 Unmodified epoxy resin ('n' denotes number of polymerized unit)

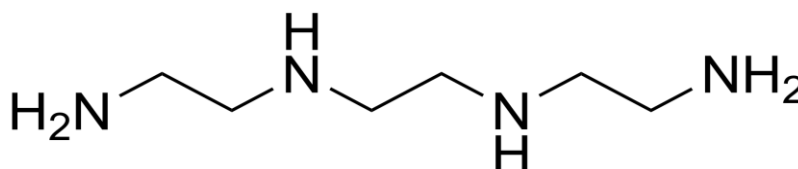


Fig. 3.2 Tri-ethylene-tetramine (hardener used for epoxy matrix)



Fig. 3.3 Epoxy resin and corresponding hardener

3.1.2 Filler material – 1: (Teak wood dust)

Teak wood dust (TWD) is produced during the cutting of teak tree and is generally treated as a biological waste with no significant utilization avenue. Teak forests are normally found in the Himalayan and sub-Himalayan sub-tropical region. The scientific name of teak is *Tectona Grandis*. It is one of three species in the genus *Tectona*. *Tectona grandis* is native to countries like India, Indonesia, Myanmar,

northern Thailand and northwestern Laos. Nilambur in Kerala (India) is a major producer of teak of fine quality and holds the world's oldest teak plantation. *Tectona grandis* is found in a variety of habitats and climatic conditions from arid areas with only 500 mm of rain per year to very moist forests with up to 5,000 mm of rain per year. Typically, though, the annual rainfall in areas where teak grows averages 1,250-1,650 mm with a 3-5 month dry season.

Table 3.2 Some important properties of Teak wood dust[77]

Characteristic Property	Inferences
Density (g/cc)	0.8
Thermal conductivity (W/mK)	0.04
Modulus of rupture (MPa)	97.1
Elastic modulus (GPa)	12.28
Tensile strength (MPa)	0.095

Main organic constituents of teak wood are: lignin, carbohydrates, hemi-cellulose, pentosans, ash, acetyl content and silica. Its dust particles appear as golden or medium brown, fibrous powder. Teak wood dust used in this work is collected from a local vendor dealing with teak furniture. The as-received teak wood dust is sieved in order to get particles in a definite size range. The particle size distribution of TWD is obtained using LASER particle size analyzer of Malvern Instruments make. Figure 3.4 shows the particle size distribution of the wood dust used in this work as the filler material. It can be seen that, majority of the dust particles are in the size range of 90-100 μ m. TWD is chosen for this work mostly for its very low thermal conductivity (0.04 W/mK) and low density (0.8gm/cc). Moreover, it is renewable, eco-friendly, available at low cost, non-toxic, its superb stability, good strength properties, easy workability and most of all, its outstanding resistance to decay and rot. It is no wonder that teak ranks among the most desired lumbers in the world. Table 3.2 provides some of the important properties. A pictorial view of teak wood dust (100 μ m) used in this present work as the particulate filler is given in Figure 3.5.

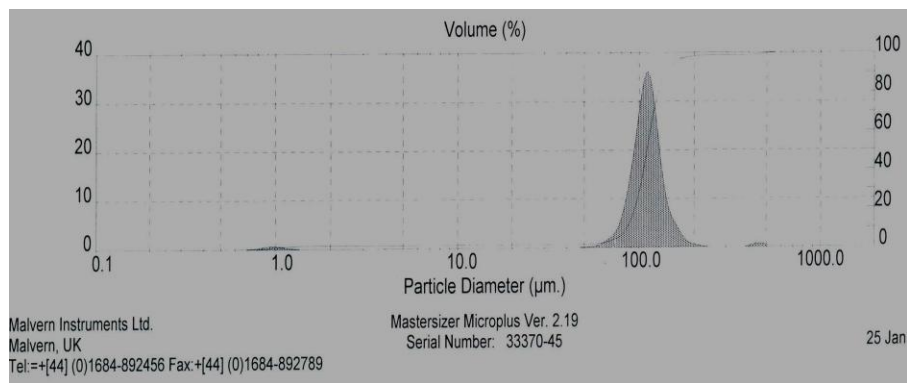


Fig. 3.4 Particle size distribution of raw TWD



Fig. 3.5 Teak wood dust



Fig. 3.6 Short banana fiber

3.1.3 Filler Material – 2: (Short Banana Fiber)

For fabrication of subsequent sets of polymer composites, a typical natural fiber i.e. banana fiber is chosen as the reinforcing material. Scientific name of banana is *musa acuminata*. At present the banana fiber is a waste product of banana cultivation, therefore without any additional cost these fibers can be obtained for industrial purpose [211]. They have a high tensile strength and resist rot. Historically they have been used to make rope. Banana fiber is a bast fiber which is a waste product of

banana cultivation. It is a ligno-cellulosic fiber, which can be extracted from the pseudo-stem of banana plant with better mechanical properties [212,213]. Main organic constituents of banana fiber are: cellulose, hemicellulose, pectin, lignin and some extractives. Banana fiber is considered to be remarkable filler because of its very low density (0.2 gm/cc), low cost, nontoxicity, biodegradability and eco-friendliness. It possesses very low thermal conductivity (0.09 W/m-K) which is the prime requirement for present investigation. Banana fibers used in present investigation are procured from M/s ROPE (Rural Opportunity Production Enterprises) International, India and are cut into pieces so as to get short banana fibers of average length 3-4 mm. Table 3.3 provides some of the important properties of banana fiber and Figure 3.6 presents a pictorial view of the short banana fibers used as reinforcement in the present work.

Table 3.3 Density, conductivity and strength properties of banana fiber [127]

Property	Values
Density	0.2 gm/cc
Thermal conductivity	0.09 W/mK
Tensile strength	49.85 MPa
Compressive strength	22.25 MPa

3.2 EXPERIMENTAL DETAILS

3.2.1 Composite Fabrication

Set 1 and Set 2: Epoxy composites filled with teak wood dust

Hand lay-up is the simplest and oldest open molding method of the composite fabrication processes. It is a low volume, labor intensive process. Room temperature curing polyesters and epoxies are the most commonly used matrix resins. Curing is initiated by a catalyst in the resin system, which hardens the fiber/filler reinforced resin composite without external heat. For a high quality part surface, a pigmented gel coat is first applied to the mold surface.

In the present work, all the epoxy based composite samples of various compositions with different fillers are prepared by this hand-lay-up technique in the following steps-

- Low temperature curing epoxy resin (LY 556) and corresponding hardener (HY 951) are mixed in a ratio 10:1 by weight as recommended.
- Teak wood dust (TWD) of average particle size 100 μm is mixed in different proportions with the resin to prepare the composites according to the experimental requirements.
- The uniformly mixed dough (epoxy filled with TWD) is then slowly decanted into the glass molds (cylindrical and rectangular shaped), coated beforehand with wax and a uniform thin film of silicone-releasing agent. Release agents facilitate easy removal of parts from molds. These products can be added to the resin, applied to molds, or both. Zinc stearate is a popular mold release agent that is mixed into resin for compression molding. Waxes, silicones and other release agents may be applied directly to the surface of molds. Release agents must be used at the lowest possible concentration. The composites are cast in different glass molds so as to get disc type specimens (diameter 40 mm, thickness 5 mm), cylindrical specimens (length 10 mm, diameter 10 mm) and rectangular slab specimens (length 150 mm, width 20 mm, thickness 3 mm).
- Composites of different compositions with varying TWD content (Table 3.4 and 3.5) are made. The castings are left to cure at room temperature for about 24 hours after which the glass molds are broken and samples are released.

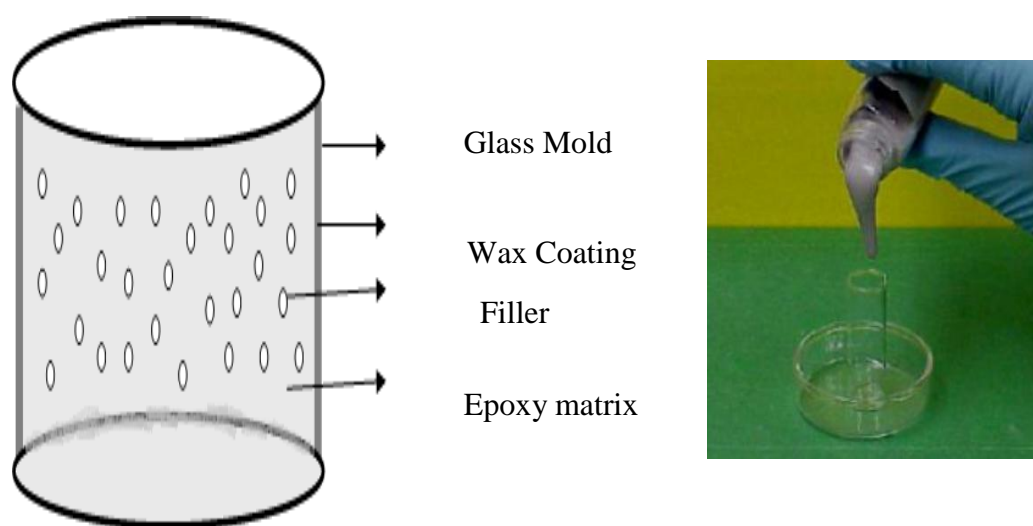
Table 3.4 Set-1 Epoxy composites filled with teak wood dust

Composition
Epoxy + 0 vol% TWD
Epoxy + 3.4 vol% TWD
Epoxy + 6.8 vol% TWD
Epoxy + 11.3 vol% TWD
Epoxy + 26.8 vol% TWD

Table 3.5 Set-2 Epoxy composites filled with Teak wood dust

Designation	Composition
ET0	Epoxy + 0 vol% TWD
ET5	Epoxy + 5 vol% TWD
ET10	Epoxy + 10 vol% TWD
ET15	Epoxy + 15 vol% TWD
ET20	Epoxy + 20 vol% TWD
ET25	Epoxy + 25 vol% TWD
ET30	Epoxy + 30 vol% TWD

A schematic diagram of the fabrication process using hand-layup technique for particulate filled epoxy composites is given in Figure 3.7. Figure 3.8 shows the composite samples of different shapes prepared for different characterization tests. Epoxy composites filled with short banana fibers (SET 3) and hybrid composites (SET 4) filled with both SBF and TWD in different proportions are fabricated following the same route.

**Fig. 3.7** Particulate filled epoxy composite fabrication by hand lay-up process

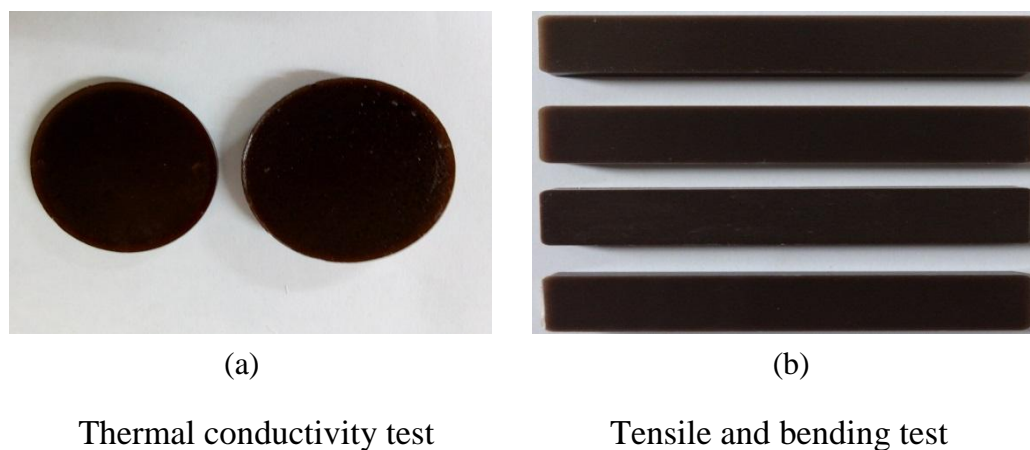


Fig. 3.8 Teak wood dust filled epoxy composites

Set 3: Epoxy Composites filled with short banana fiber (SBF)

As already mentioned, using the same method, short banana fibers with an aspect ratio of about 150 are reinforced in different proportions into the epoxy resin to prepare the composites as listed out in Table 3.6. The composites are cast into glass molds so as to get disc type and rectangular slab type specimens. A typical disc shaped epoxy-SBF composite is shown in Figure 3.9.

Table 3.6 Set-3 Epoxy composites filled with short banana fibers

Designation	Composition
EB0	Epoxy + 0 vol% SBF
EB5	Epoxy + 5 vol% SBF
EB10	Epoxy + 10 vol% SBF
EB15	Epoxy + 15 vol% SBF
EB20	Epoxy + 20 vol% SBF



Fig. 3.9 Shortbanana fiber reinforced epoxy composite

Set 4: Epoxy Composites filled with hybrid fillers (TWD and SBF)

Further, a set of hybrid composites are fabricated so as to study the synergistic effects of reinforcement of multiple fillers on various composite properties. Composites of six different compositions with varying TWD and SBF contents are included in this set of hybrid specimens. The TWD content is varied from 10 to 20 vol% and the SBF content is varied from 0 to 10 vol%. Table 3.7 shows the detailed compositions of the hybrid samples prepared for this investigation and Figure 3.10 illustrates a typical epoxy-TWD-SBF hybrid composite.

Table 3.7 Set-4Epoxy composites filled with hybrid fillers

Designation	Composition
ETB1	Epoxy + 10 vol% TWD + 0 vol% SBF
ETB2	Epoxy + 10 vol% TWD + 5 vol% SBF
ETB3	Epoxy + 10 vol% TWD + 10 vol% SBF
ETB4	Epoxy + 20 vol% TWD + 0 vol% SBF
ETB5	Epoxy + 20 vol% TWD + 5 vol% SBF
ETB6	Epoxy + 20 vol% TWD + 10 vol% SBF



Fig 3.10 TWD and SBF based hybrid composite

3.3 COMPOSITE CHARACTERIZATION

3.3.1 Physical Characterization

Density and void fraction

The densities of composite materials and their components are normally measured using one of three methods: The Archimedes method, the sink-float method, or the density gradient method.

In the present work, the actual density (ρ_a) of composite is determined by the Archimedes principle using distilled water as the medium. This method is covered in ASTM standard D 792. According to this principle when an object is immersed in a liquid the apparent loss in its weight is equal to the up thrust and this is equal to the weight of the liquid displaced. The density of the composite is obtained by using equation 3.1.

$$\rho_a = \frac{\rho_w W_a}{W_a - W_w} \quad (3.1)$$

Here ρ_a is the actual/measured density of composite, ρ_w is the density of distilled water, W_a is weight of the sample in air and W_w is weight of the sample in water. The theoretical density of composite materials (ρ_t) in terms of weight fraction can easily be obtained as per the following equation given by Agarwal and Broutman [214].

$$\rho_t = \frac{1}{\left(\frac{W_p}{\rho_p}\right) + \left(\frac{W_m}{\rho_m}\right)} \quad (3.2)$$

Where, W and ρ represent the weight fraction and density respectively. The suffix p and m stand for the particulate filler and matrix material respectively.

In the present work, since some of the composites are hybrid type i.e filled with double fillers (TWD and SBF), the above expression for density has been modified as:

$$\rho_t = \frac{1}{\left(\frac{W_f}{\rho_f}\right) + \left(\frac{W_m}{\rho_m}\right) + \left(\frac{W_p}{\rho_p}\right)} \quad (3.3)$$

Where, the suffix ' f ' indicates the fiber.

The presence of voids will add to the total volume, but not the weight of the composite. V_v is the void content which is then expressed as:

$$V_v = \frac{(\rho_t - \rho_a)}{\rho_t} \quad (3.4)$$

Scanning Electron Microscopy

The micro-structural features of the various particulate filled and fiber reinforced composite specimens are examined by Scanning Electron Microscope JEOL JSM-6480 LV (Figure 3.11). The specimens are cleaned, dried and mounted on stubs with silver paste. To improve the penetration of light and for better surface micrographs, a thin film of platinum is vacuum-evaporated onto the samples before the micro-graphs are taken.

3.3.2 Mechanical Characterization

Tensile Strength

Tensile strength of any composite indicates the maximum axial pull it can withstand without failure. The cured rectangular shaped composite samples of required dimension (length 150mm, width 20mm and thickness 3mm) are used for the tensile

test as per ASTM E 1309 standard. A uniaxial load is applied through both the ends. In the present work, this test is performed in the universal testing machine Instron 1195 (Figure 3.12) at a cross head speed of 10 mm/minute and the results are used to calculate the tensile strength of the composite samples. The loading arrangement is shown in Figure 3.13a. Typical rectangular shaped samples used for tensile test are shown in Figure 3.8b. In this case, the test is repeated three times on each composite type and the mean value is reported as the tensile strength of that composite.

Compressive strength

Compressive strength of any composite indicates the maximum axial compressive load it can withstand without failure. The cured composite samples of required dimension (length 10mm, diameter 10mm) are used for the uniaxial compression test as per ASTM D 695. This helps to avoid buckling, reduce friction due to small cross-sectional area, avoid premature failure due to sharp corners and prevent self-reaction of the resin during curing. Static uniaxial compression tests are carried out on the specimens using the same UTM *Instron1195*. The loading arrangement is shown in Figure 3.13b. Three identical specimens are tested for each composition and the average result is recorded as the compressive strength. Crosshead speed is maintained at 1mm/min and the compression is stopped when the specimen shows signs of failure or when densification is complete.

Flexural Strength (FS)

The flexural strength of a composite is the maximum tensile stress that it can withstand during bending before reaching the breaking point. The three point bend test is conducted on all the composite samples as per ASTM D 7264 in the universal testing machine Instron 1195 for evaluating flexural strength. The dimension of each specimen is 60 mm × 10 mm × 3 mm. Span length of 40 mm and the cross head speed of 10 mm/min are maintained. The test is repeated three times for each composite type and the mean value is reported. The flexural strength of the composite specimen is determined using the following equation:

$$F.S. = \frac{3PL}{2bt^2} \quad (3.5)$$

Where, L is the span length of the sample (mm), P is maximum load (N), b is the width of the specimen (mm) and t is the thickness of the specimen (mm).



Fig. 3.11 Scanning Electron Microscope (JEOL JSM-6480LV)



Fig. 3.12 Universal testing machine *Instron 1195*

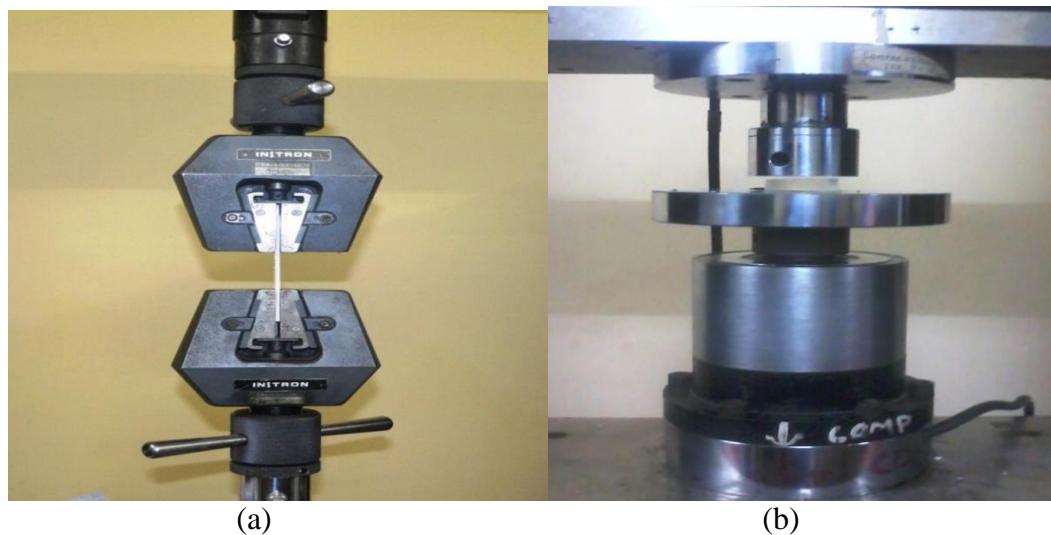


Fig. 3.13 (a) Loading arrangement for tensile test
(b) Loading arrangement for compression test

3.3.3 Thermal Characterization

Thermal Conductivity: Experimental Determination

Equipment used: UnithermTM Model 2022 Thermal Conductivity Tester

Test standard: ASTM E 1530

Operating Principle

By definition “Thermal conductivity is the exchange of energy between adjacent molecules and electrons in a conducting medium, it is a material property that describes heat flow within a body for a given temperature difference per unit area.”

For one dimensional heat flow, the equation is given as:

$$Q = kA \left(\frac{T_1 - T_2}{x} \right) \quad (3.6)$$

Where, Q is the heat flux (W), A is the cross-sectional area (m^2), k is thermal conductivity (W/mK), x is the sample thickness (m), $T_1 - T_2$ is the temperature difference between surfaces ($^{\circ}\text{C}$ or K). The thermal resistance of the sample is given as:

$$R = \frac{T_1 - T_2}{Q} \quad (3.7)$$

Where, R is sample resistance between hot and cold surfaces. (K/W) From the former equation, we can write:

$$k = \frac{x}{RA} \quad (3.8)$$

In UnithermTM 2022, transducers measure the value of heat flux Q and temperature difference between upper and lower plate. Thus, thermal resistance between surfaces can be evaluated. Providing different thickness and known cross-sectional area as input parameters, the sample thermal conductivity can be calculated.

Glass transition temperature and Coefficient of thermal expansion: Experimental Determination

Equipment used: Thermal Mechanical Analyzer

Glass transition temperature (T_g) is the temperature at which the mechanical properties of amorphous polymer changes from the brittle state of glass to a rubbery state. The most important property change at the glass transition temperature is the thermal expansion, which is a swift from a relatively low value to a very high value. This kind of change is not desirable as it imposes stress when they experience high-temperatures during manufacturing, assembly or during their service-life. Similarly, coefficient of thermal expansion (CTE) is the rate of change of the expansion of a material with temperature.



Fig. 3.14 Thermal Conductivity Tester *Unitherm™* 2022



Fig. 3.15 Perkin Elmer DSC-7 Thermal Mechanical Analyzer

In the present work, T_g and CTE of the composites are measured with a *Perkin Elmer DSC-7 Thermal Mechanical Analyzer* (TMA) in expansion mode (Figure 3.15). At first, the TMA sample stage is purged with nitrogen gas. The sample length is set between 6-8 mm and the width as well as thickness was about 2-3 mm. During the TMA measurement, the specimen is heated from 30 to 150°C at a heating rate of 5°C/min. For each measurement, two heating scans are used. The first heating scan is used to eliminate any possible internal stress and moisture in the sample which is likely to be generated during the curing and sample preparation process. The second heating scan is used to determine the T_g and CTE of the material.

Chapter Summary

This chapter has provided:

- The descriptions of materials (matrix and fillers) used in this research
- The details of fabrication of the composites
- The details of physical, mechanical and thermal characterization tests.

The next chapter presents the test results related to the physical and mechanical properties of the polymer composites under this study.

CHAPTER 4

Results and Discussion – I

Physical and Mechanical Characteristics Of the Composites

Chapter 4**Results and Discussion – I****PHYSICAL AND MECHANICAL CHARACTERISTICS
OF THE COMPOSITES**

This chapter presents the measured values of the physical and mechanical properties of the epoxy composites filled with micro-sized teak wood dust (TWD) and/or short banana fibers (SBF). The effects of these particulate and fiber fillers on various physico-mechanical properties of epoxy composites have been discussed. It also presents some of the scanning electron micrographs showing typical micro-structural features of various composites under this study.

4.1 PHYSICAL CHARACTERISTICS**4.1.1 Density and volume fraction of voids**

The theoretical and measured densities along with the corresponding volume fraction of voids in the epoxy-TWD and epoxy-SBF composites are presented in Tables 4.1 and 4.2 respectively. Similarly Table 4.3 presents these values for the set of hybrid composites i.e. epoxy filled with pre-determined proportions of both TWD and SBF. It may be noted that the composite density values calculated theoretically from weight fractions using Eqn.3.2 and Eqn. 3.3 are not equal to the experimentally measured values. This difference is a measure of voids and pores present in the composites. It is found that with the increase in TWD content in epoxy resin from 0 to 40 vol%, there is a drop in density of the composite by about 15 % although there is a simultaneous increase in the void fraction or porosity from 0.733% to 5.603% (Table 4.1). Similarly, a substantial drop in composite density by about 34.80% is observed as the SBF content in epoxy is increased from 0 to 20 vol% (Table 4.2). Reasonable

decrement in density is also noted in case of all the hybrid composites under this study (Table 4.3).

Table 4.1 Measured and Theoretical densities of the composites
(Epoxy filled with TWD)

Composite Designation	Filler (TWD) Content		Measured Density (gm/cc)	Theoretical Density (gm/cc)	Volume Fraction of Voids (%)
	vol %	wt%			
ET0	0	0	1.092	1.100	0.733
ET5	5	3.69	1.071	1.085	1.307
ET10	10	7.47	1.060	1.070	0.943
ET15	15	11.37	1.042	1.055	1.248
ET20	20	15.40	1.024	1.039	1.465
ET25	25	20.30	1.008	1.022	1.389
ET30	30	23.76	0.992	1.010	1.815
ET35	35	28.14	0.976	0.995	1.947
ET40	40	32.65	0.928	0.980	5.603

Table 4.2 Measured and Theoretical densities of the composites
(Epoxy filled with SBF)

Composite Designation	Filler (SBF) Content		Measured Density (gm/cc)	Theoretical Density (gm/cc)	Volume Fraction of Voids (%)
	vol %	wt%			
EB0	0	0	1.089	1.100	1.010
EB5	5	0.95	1.042	1.055	1.248
EB10	10	1.98	0.970	1.010	4.124
EB15	15	3.11	0.910	0.965	6.044
EB20	20	4.35	0.710	0.774	9.014

Table 4.3 Measured and Theoretical densities of the hybrid composites
(Epoxy filled with TWD and SBF)

Composite Designation	TWD Content (vol%)	SBF Content (vol%)	Measured Density (gm/cc)	Theoretical Density (gm/cc)	Volume Fraction of Voids (%)
ETB1	10	0	1.055	1.070	1.422
ETB2	10	5	1.014	1.027	1.282
ETB3	10	10	0.971	0.985	1.442
ETB4	20	0	1.022	1.040	1.761
ETB5	20	5	0.977	0.999	2.252
ETB6	20	10	0.914	0.927	1.422

Density of a composite depends on the relative proportion of matrix and reinforcing materials and this is one of the most important factors determining the properties of the composites. The void content is the cause for the difference between the values of true density and the theoretically calculated ones. The voids significantly affect some of the mechanical properties and even the performance of the composites in the workplace. Higher void content usually mean lower fatigue resistance, greater susceptibility to water penetration and weathering [214]. The knowledge of void content is desirable for better estimation of quality of the composites. It is understandable that a good composite should have fewer voids. However, presence of void is unavoidable particularly in composites made through hand-lay-up route.

4.1.2 Micro-structural characterization

It is well known that the properties of the composites are strongly dependent on the interaction between the filler and the matrix. In order to evaluate this interaction, the microstructure of the composites including the dispersion of the micro-sized teak wood dust particles in the epoxy matrix is observed under scanning electron microscope. Figure 4.1a shows a typical SEM image of TWD filled epoxy composite.

A single TWD particle (within the matrix body) at a higher magnification is shown in Figure 4.1b. These micrographs confirm the near- spherical shape of the filler particles i.e. TWD used as reinforcement in the polymer resin. The surface morphologies of epoxy composites reinforced with short banana fibers are also shown in Figures 4.2.a and b. These micrographs exhibit a peculiar arrangement of bony ridges of different relative size and thickness on the fiber body embedded in the epoxy resin.

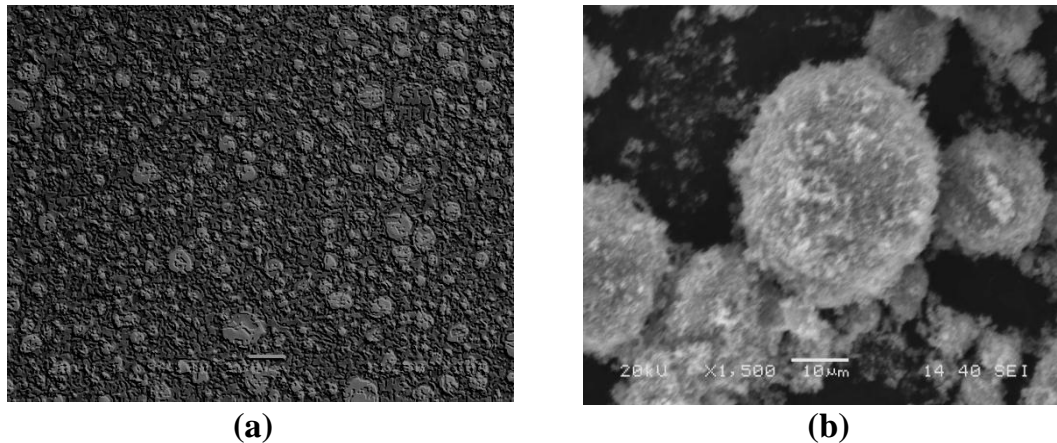


Fig. 4.1 SEM images of epoxy composites filled with micro-sized TWD particles

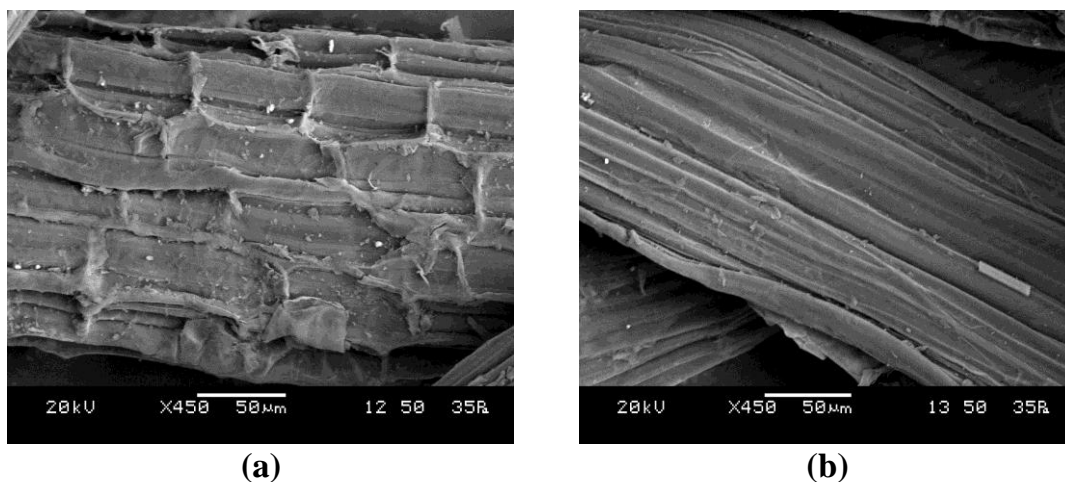


Fig. 4.2 Some typical SEM images of short banana fibers

4.2 MECHANICAL CHARACTERISTICS

A wealth of property data has been generated by conducting different characterization tests under controlled laboratory conditions to evaluate various mechanical characteristics of the composites fabricated for this work.

4.2.1 Tensile Strength

Tensile strengths of the composite specimens are evaluated and the test results for the epoxy-TWD, epoxy-SBF and epoxy-TWD-SBF hybrid composites are presented in Tables 4.4, 4.5 and 4.6 respectively. It is noticed that with addition of TWD particles, tensile strength of the composite decreases and this decrement is a function of the filler content.

Table 4.4 Mechanical properties of the composites
(Epoxy filled with Teak Wood Dust)

Composite Designation	Filler (TWD) Content		Tensile Strength (MPa)	Flexural Strength (MPa)	Compressive Strength (MPa)
	vol %	wt%			
ET0	0	0	65.00	58.00	90.00
ET5	5	3.69	64.50	57.70	88.20
ET10	10	7.47	63.70	57.20	86.80
ET15	15	11.37	62.80	56.60	85.50
ET20	20	15.40	61.70	55.80	84.60
ET25	25	20.30	60.30	55.30	83.40
ET30	30	23.76	58.90	54.60	82.30
ET35	35	28.14	57.60	53.70	81.50
ET40	40	32.62	56.20	53.00	80.70

It is noted that while the tensile strength of neat epoxy is 65 MPa, it decreases by about 13.8 % to 56.2 MPa with the incorporation of 40 vol% of TWD in it. On the contrary, in case of epoxy-SBF composites, the tensile strength is found to be increasing with SBF content quite substantially. While the tensile strength of unreinforced epoxy is 65 MPa, with incorporation of 20 vol% of short banana fibers, the tensile strength reaches a value as high as 138 MPa and this increment is about 112%. Measured tensile strength values of epoxy-TWD-SBF hybrid composites with different filler and fiber proportions are also graphically illustrated in Figure 4.3 which shows a gradual increase in tensile strength values with increase in SBF proportion irrespective of the TWD content.

Table 4.5 Mechanical properties of the composites
(Epoxy filled with Short Banana Fiber)

Composite Designation	Filler (SBF) Content		Tensile Strength (MPa)	Flexural Strength (MPa)	Compressive Strength (MPa)
	vol %	wt%			
EB0	0	0	65.00	58.00	90.00
EB5	5	0.95	97.00	84.00	98.00
EB10	10	1.98	111.00	100.00	109.00
EB15	15	3.10	127.00	118.00	123.00
EB20	20	4.35	138.00	131.00	132.00

Table 4.6 Mechanical properties of the hybrid composites
(Epoxy filled with both Teak Wood Dust and Short Banana Fiber)

Composite Designation	TWD Content (vol%)	SBF Content (vol%)	Tensile Strength (MPa)	Flexural Strength (MPa)	Compressive Strength (MPa)
ETB1	10	0	63.70	57.20	86.80
ETB2	10	5	80.50	72.40	92.88
ETB3	10	10	96.40	88.45	99.76
ETB4	20	0	61.70	55.80	84.60
ETB5	20	5	76.80	68.86	90.34
ETB6	20	10	92.40	83.54	95.68

4.2.2 Compressive Strength

Compressive strengths of the composite specimens are evaluated by conducting uni-axial compression test and the test results for the epoxy-TWD, epoxy-SBF and epoxy-TWD-SBF composites are presented in Tables 4.4, 4.5 and 4.6 respectively. It is noticed that with addition of TWD particles, compressive strength of the composite

decreases and this decrement is a function of the filler content. While the compressive strength of neat epoxy is 90 MPa, it decreases by about 10 % to 80.70 MPa with the incorporation of 40 vol% of TWD in it. On the contrary, in the case of epoxy-SBF composites the compressive strength is found to be increasing with SBF content significantly. With incorporation of 20 vol% of short banana fibers the compressive strength of the composite reaches 132 MPa which is about 46.7% higher than that of the unreinforced epoxy. Measured compressive strength values of epoxy-TWD-SBF hybrid composites with different filler and fiber proportions are also graphically illustrated in Figure 4.4 which shows a gradual increase in compressive strength with increase in SBF content irrespective of the volume percentage of TWD.

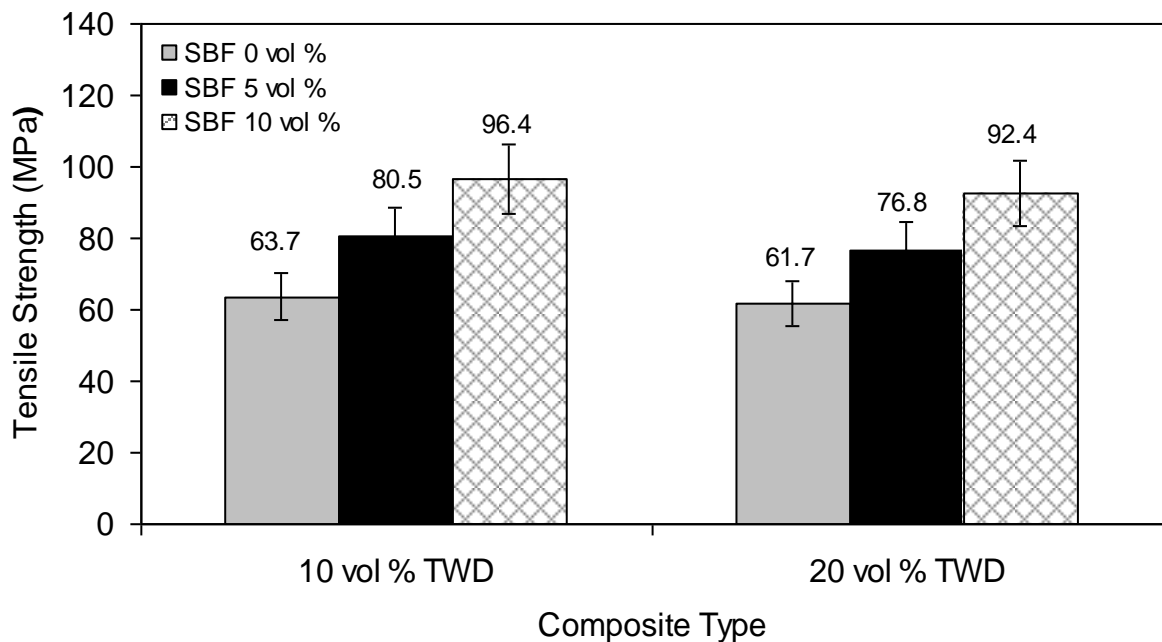


Fig. 4.3 Tensile strength of hybrid composites with different fiber and filler concentrations

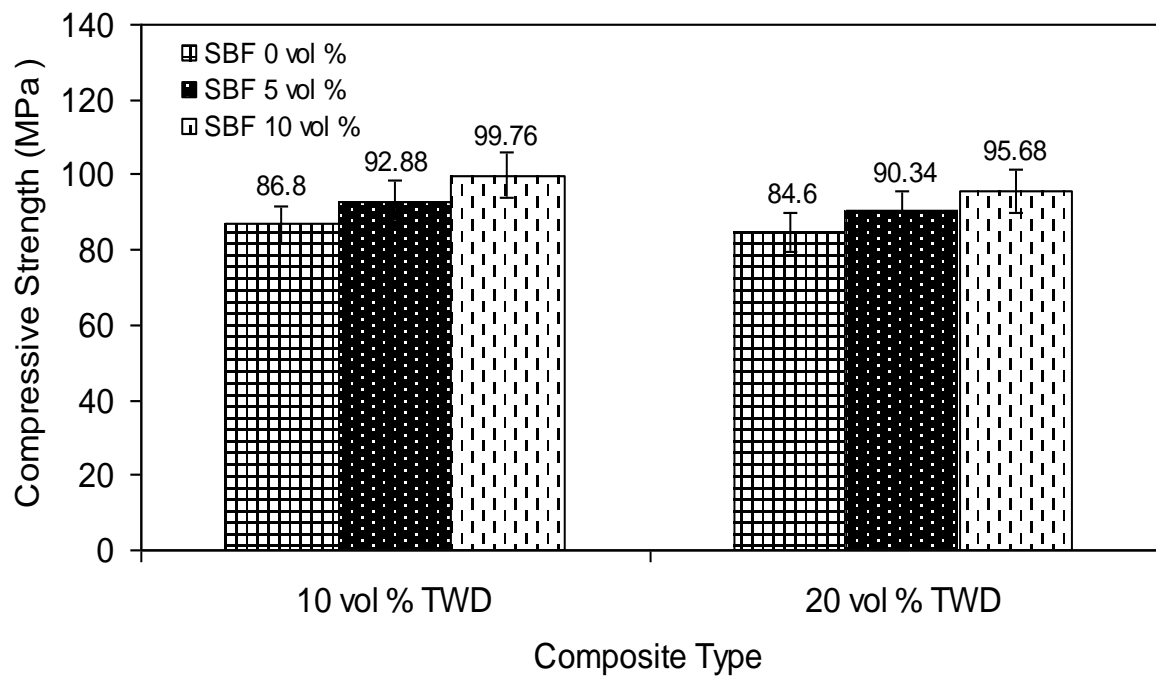


Fig. 4.4 Compressive strength of hybrid composites with different fiber and filler concentrations

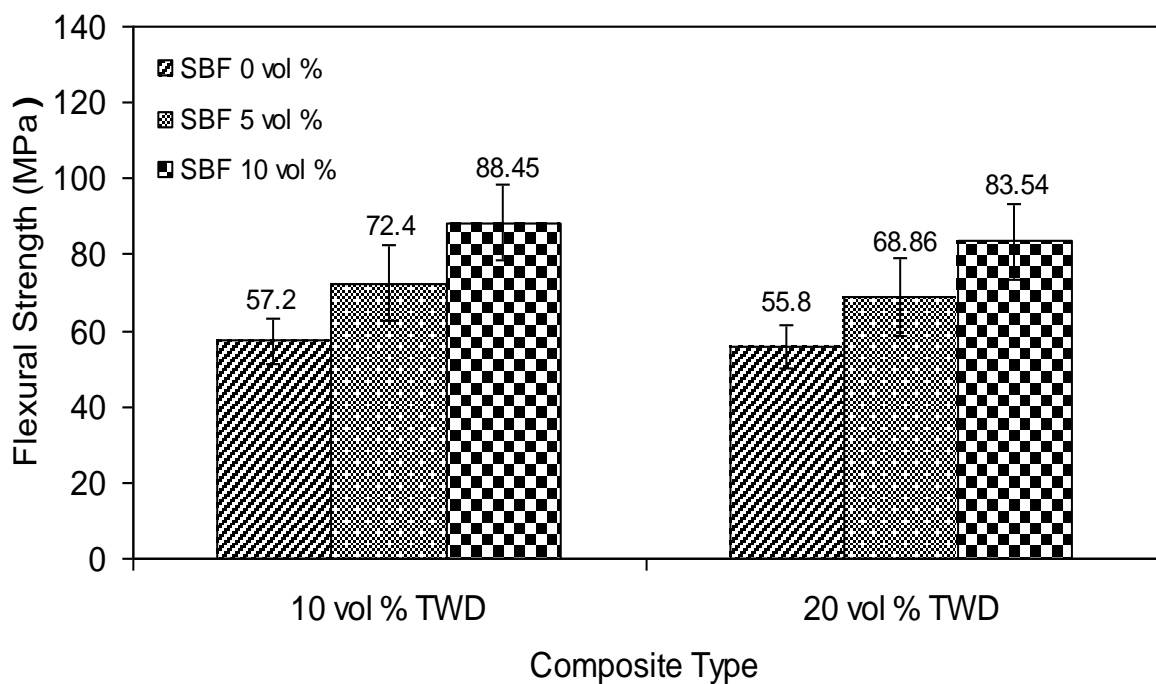


Fig. 4.5 Flexural strength of hybrid composites with different fiber and filler concentrations

4.2.3 Flexural Strength

Composite materials used in structures are prone to fail in bending and therefore development of new composites with improved flexural characteristics is essential. In the present work, flexural strengths of the composite specimens are evaluated by conducting three point bend test in accordance with ASTM D7264 and the test results for the epoxy-TWD, epoxy-SBF and epoxy-TWD-SBF composites are presented in Tables 4.4, 4.5 and 4.6 respectively. It is noticed that flexural strength of the composite decreases with addition of TWD particles whereas with reinforcement of SBF there is appreciable increase in the value of flexural strength. Measured flexural strength values of epoxy-TWD-SBF hybrid composites with different filler and fiber proportions are also graphically illustrated in Figure 4.5 which shows a gradual increase in flexural strength values with increase in SBF content irrespective of the volume percentage of TWD in it.

Discussion

Density values of all the particulate or short fiber filled composites under this study are found to be lower than that of the matrix polymer since both teak wood dust and short banana fiber possess much lower densities than that of epoxy. Mechanical characterization of the composites under this investigation reveals that the trends observed in respect to the strength properties for TWD filled composites are quite different from those obtained in the case of SBF reinforced epoxy composites. There can be two reasons for the reduction of various strength properties as the teak wood dust content increases. The first reason may be due to the fact that the chemical reaction at the interface between the filler particle and the matrix is too weak to transfer the tensile stress; the second reason may be due to the rise of the stress concentration at the sharp corner of some of the irregularly shaped particulates embedded in the epoxy matrix. However this drop in strength can be compensated and strength can further be enhanced by additional reinforcement of short fibers in the epoxy resin.

Chapter Summary

Several important conclusions emerge from the extensive results obtained in these studies carried out on epoxy-TWD, epoxy-SBF and epoxy-TWD-SBF hybrid composites in regard to their physical, mechanical and micro-structural characteristics. The results presented in this chapter clearly indicate a trendy characterization in the behavior of different particulate/short-fiber filled composites under this study and they provide a very important criterion for the choice of the composites for specific needs.

The next chapter presents the test results related to the thermal characteristics of the composites filled with micro-sized teak wood dust particles and/or short banana fibers with emphasis on their thermal insulation capabilities.

CHAPTER 5

Results and Discussion – II

Thermal CharacteristicsOf the Composites

Chapter 5

Results and Discussion – II

THERMAL CHARACTERISTICS OF THE COMPOSITES

This chapter presents the test results for thermal properties such as effective thermal conductivity (k_{eff}), glass transition temperature (T_g) and coefficient of thermal expansion (CTE) of all the epoxy-TWD, epoxy-SBF, epoxy-TWD-SBF composites under this investigation. A theoretical one-dimensional heat conduction model has been developed and based on this, a correlation for the estimation of effective thermal conductivity of polymer composites with micro-sized particulate fillers has been proposed. The results of the numerical analysis and validation of the proposed theoretical model in regard to the determination of effective thermal conductivity are presented in this part of the thesis. The effects of particulate fillers and short fibers on T_g and CTE of the composites are also discussed.

5.1. EFFECTIVE THERMAL CONDUCTIVITY (k_{eff})

Effective thermal conductivities of epoxy composites filled with TWD fillers are evaluated theoretically and experimentally. The interpretation and comparison of results obtained from different methods for composites with different filler concentrations are presented.

5.1.1. Numerical Methods: Concept of Finite Element Method and ANSYS

The numerical simulations have been used for decades in order to explore the mechanism of thermal conduction in composites and finite element method (FEM) has been considered as the most powerful tool among them. In general, FEM requires the generation of a new model for every special composite system to be explored. However, it is laborious to create model for each specific issue. To overcome the

problems associated with model creation, ANSYS, a mature means of FEM is usually chosen. In order to evaluate the effective properties of composite, the finite element software package ANSYS is used. The program is written in APDL (ANSYS Programming Design Language), which is delivered by the software and it makes the handling much more comfortable. The finite element method (FEM), originally introduced by Turner et al. [215], is a powerful computational technique for approximate solutions to a variety of “real-world” engineering. FEM is considered one of the most promising methods for studying the thermal conductivity (TC) enhancement/reduction of composite materials. This method has already been used to predict the TC of a wide range of two-phase composites with many effects, including Cu/PP, AlN/PI [216] and nanocomposites based on epoxy [217].

Ramani and Vaidyanathan [218] established an automated finite element analysis method to determine the effective thermal conductivity of composites. A few investigations on thermal conductivities of some filled polymer composites using numerical and experimental methods have also been conducted recently [219-221]. The FEM is a numerical procedure that can be used to obtain solutions to a large class of engineering problems involving stress analysis, heat transfer, fluid flow etc. ANSYS is a general purpose finite-element modeling package for numerically solving a wide variety of mechanical problems that include static/dynamic, structural analysis (both linear and nonlinear), heat transfer and fluid problems, as well as acoustic and electromagnetic problems.

The basis of FEM relies on the discretization of the solution domains into finite elements for which systematic approximate solution is constructed by applying the variational or weighted residual methods. In effect, FEM reduces the problem to that of a number of unknowns by dividing the domains into elements and by expressing the unknown field variable in terms of the assumed approximating functions within each element. These interpolation functions are defined in terms of the values of the field variables at specific points referred as nodes. Nodes are usually located along the element boundaries and they connect adjacent elements. The ability to discretize the irregular domains with finite elements makes the method a valuable and practical

analysis tool for the solution of boundary, initial and eigenvalue problems arising in various engineering disciplines.

Basic Steps in Finite Element Method

Solving a general engineering problem in FEM involves the following steps:

- The first step in the finite element method is to divide the structure or continuum into subdivisions or elements. Then the structure is to be modeled with suitable finite elements. The size, shape, arrangement and number of elements are to be decided in the first step.
- In the second step, a proper interpolation model is to be selected. Since the solution of a complex structure under any specified conditions cannot be predicted exactly, a suitable solution within an element is assumed to approximate the unknown solution. The assumed solution must be simple from a computational standpoint, but it should satisfy certain convergence requirements.
- In the third step, element characteristic matrices and input data are to be derived from the assumed interpolation model by using either equilibrium conditions or a suitable vibrational principle.

Description of the problem:

The estimation of effective properties of composites is of vital importance for proper design and application of composite materials. The micro-structural characteristics are essential criteria which influence the effective properties of the composite. Micro-structure means the shape, size, spatial distribution and orientation of the embedment within the matrix. Despite the fact that most composite systems have inclusions of random distribution and orientation, a great insight of the effect of microstructure on the effective properties can be achieved from the analysis of composites with periodic structure. A system with high degree of symmetry in its structure is preferred as it can be easily analyzed. Based on micro-structure, composites can generally be classified as laminate composite, i.e. with multi-layered structure and dispersion composite, i.e.

including a matrix phase and one or more dispersion phases. In this work, models of dispersion composites are generated. The factors affecting the effective thermal conductivity of dispersion composites are structural features of the composite and thermal conductivity of each constituent. The structural features include shape and size of matrix, volume fraction, shape, size and orientation of dispersion. The structure of dispersion composite is considered to be composed of some very simplified basic models. In a basic model, the shape of composite is usually assumed to be cubical.

In the present investigation, micro-sized teak wood dust particles are used as the filler material embedded in epoxy resin. A typical scanning electron microscope (SEM) image of a single TWD particle is shown in Figure 5.1 which indicates the filler shape to be nearly spherical. Hence, the FEM models considered for this analysis include cubical shaped epoxy matrix with spherical inclusions.

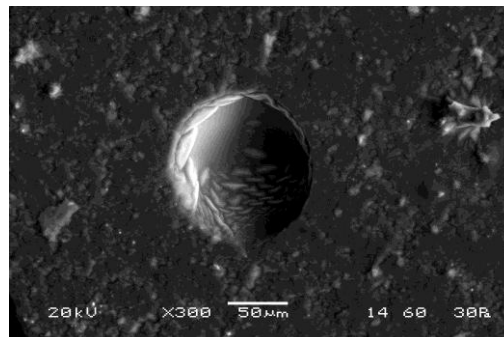


Fig. 5.1 A typical SEM image of TWD particle inside the matrix

A schematic representation of TWD particles embedded in an epoxy body having a periodic arrangement and the prescribed boundary conditions with the direction of heat flow for the conduction problem is shown in Figure 5.2. The temperature at the nodes along the surface ABCD is T_1 ($=100^\circ\text{C}$), the ambient convective heat transfer coefficient is assumed to be $2.5 \text{ W/m}^2\text{K}$ and the analysis of the problem is done at a room temperature of 27°C . The other surfaces parallel to the direction of the heat flow are assumed to be adiabatic. The temperatures at the inside domain and on the other boundaries are not apprehended. A few presumptions involved in this analysis are:

- The composites are macroscopically homogeneous.
- Locally both the matrix and filler are homogeneous and isotropic.
- The filler-matrix interfacial thermal contact resistance is negligible.
- The composite body is free from voids.
- The fillers are assumed to be uniformly distributed in the matrix in a square periodic array.

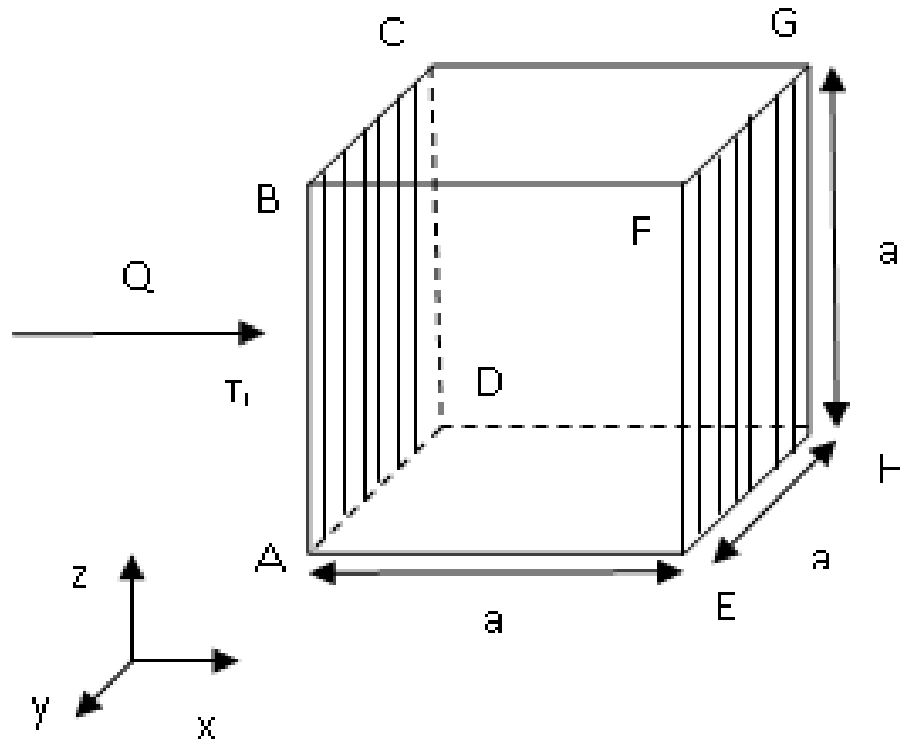
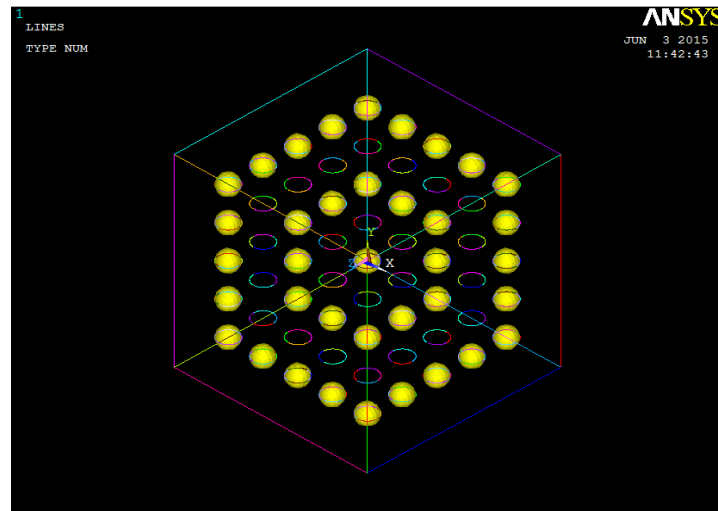
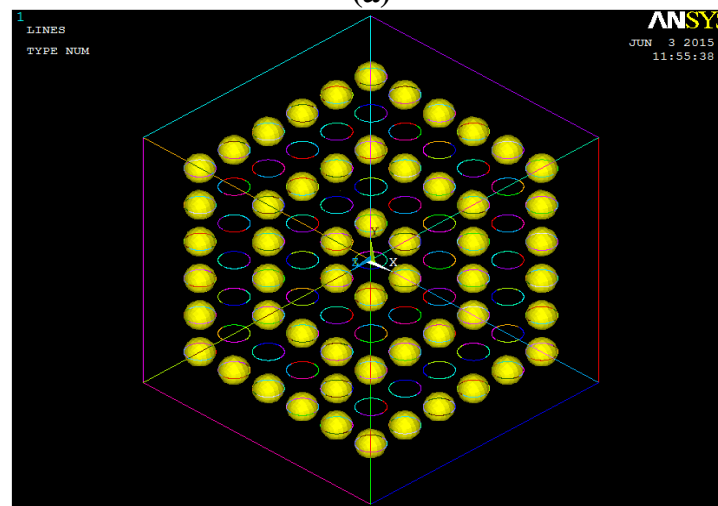


Fig. 5.2 The heat flow direction and boundary conditions for the particulate-polymer composite

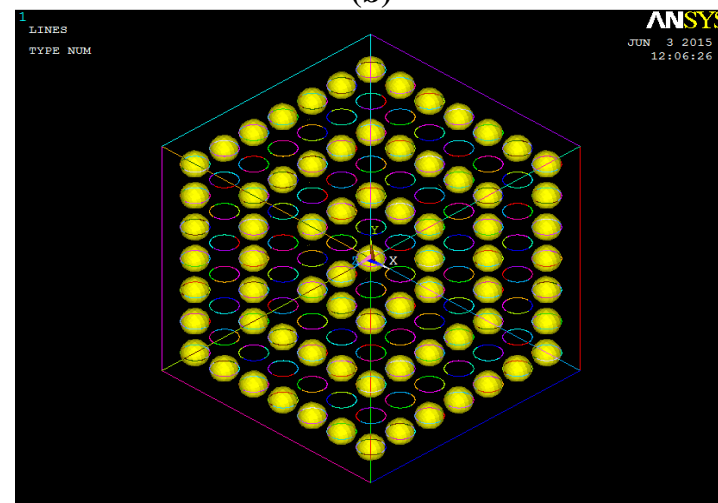
Thermal conductivities of the TWD-epoxy composites are numerically estimated by using the sphere-in-cube model. Some typical 3-D physical models showing the arrangement of TWDmicro spheres within the cube shaped composites with filler concentrations of 6.5 vol%, 11.3 vol% and 17.95 vol% are illustrated in Figures 5.3a, 5.3b and 5.3c respectively. Similarly, the temperature profiles obtained from FEM analysis for composites with TWD concentrations of 1.41, 3.4, 6.5, 11.3, 17.95 and 26.8 vol% are presented in Figures 5.4a, 5.4b, 5.4c, 5.4d, 5.4e and 5.4f respectively.



(a)

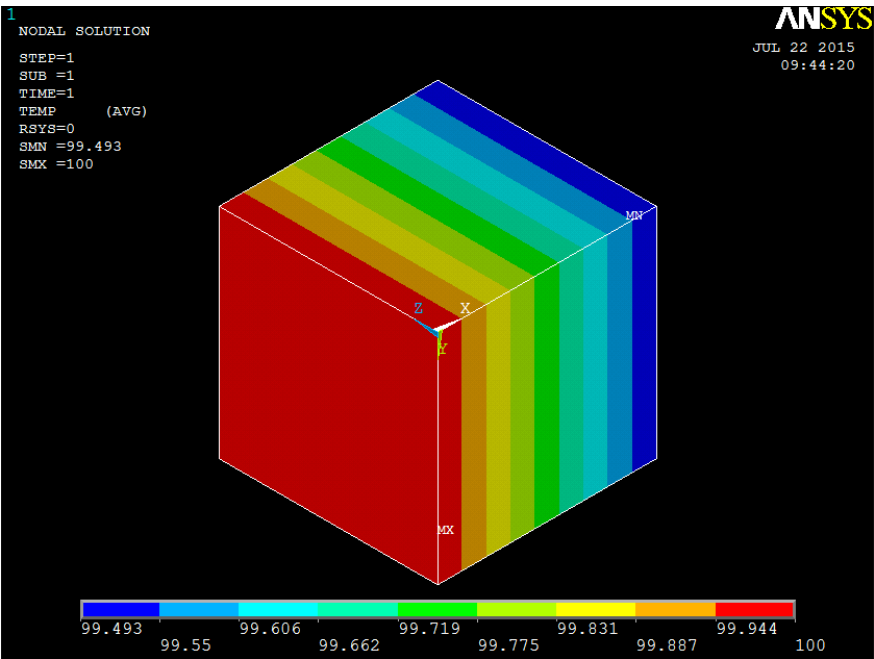


(b)

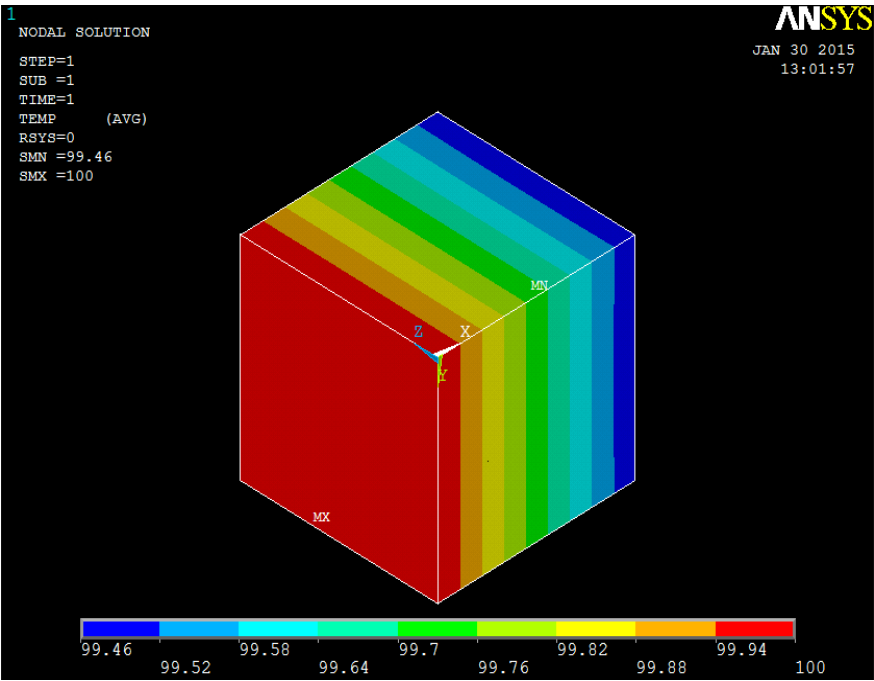


(c)

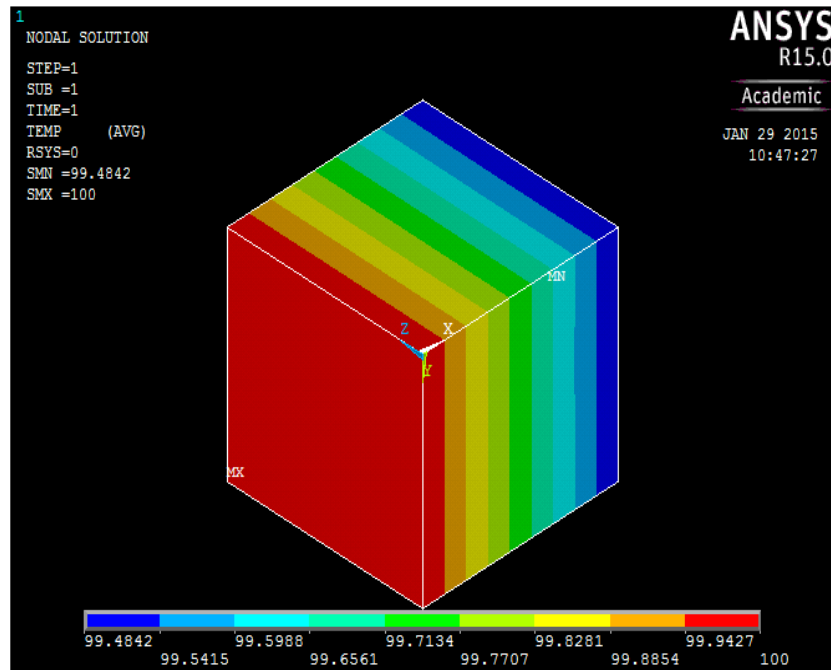
Fig. 5.3 Typical 3-D sphere-in-cube models for composites with different filler concentrations (a) 6.5 vol% (b) 11.3 vol% and (c) 17.95 vol%



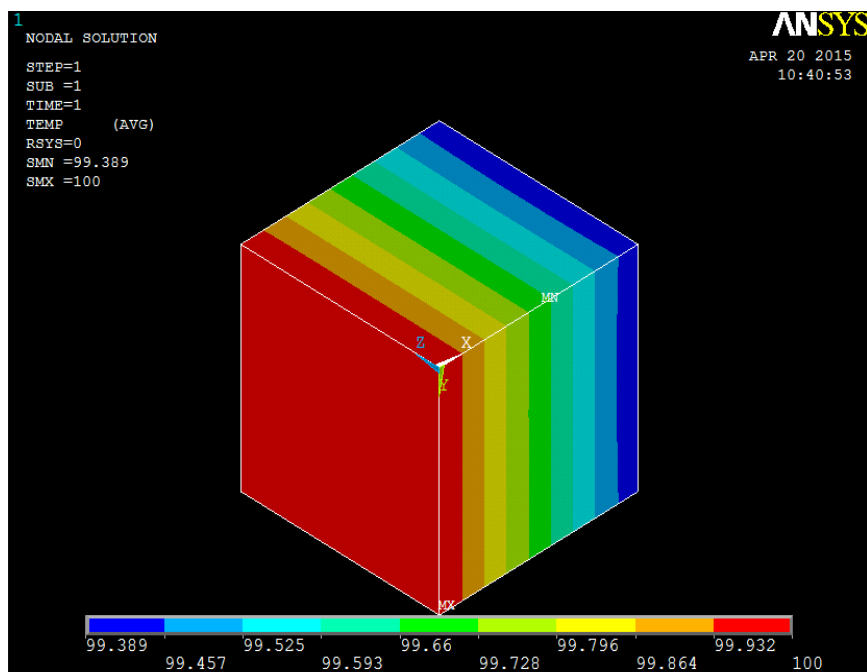
(a)



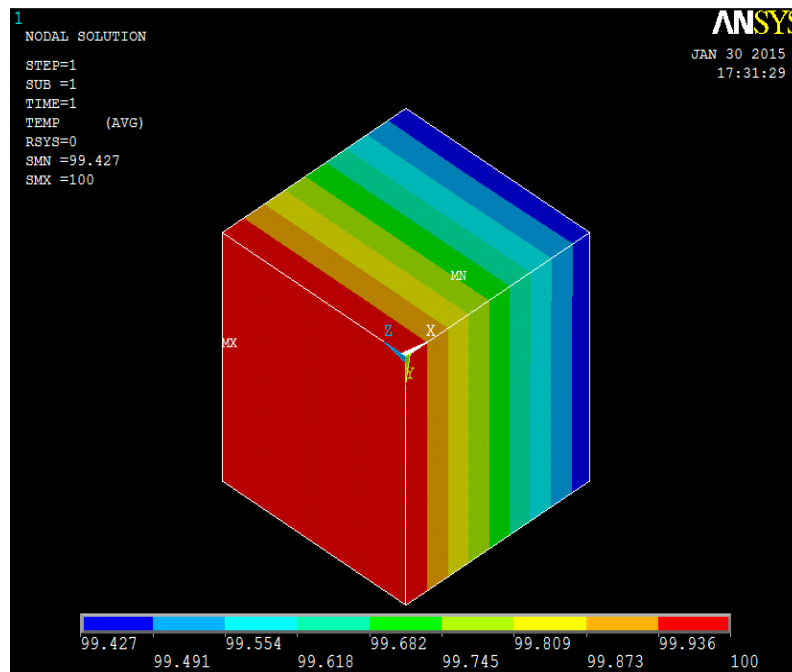
(b)



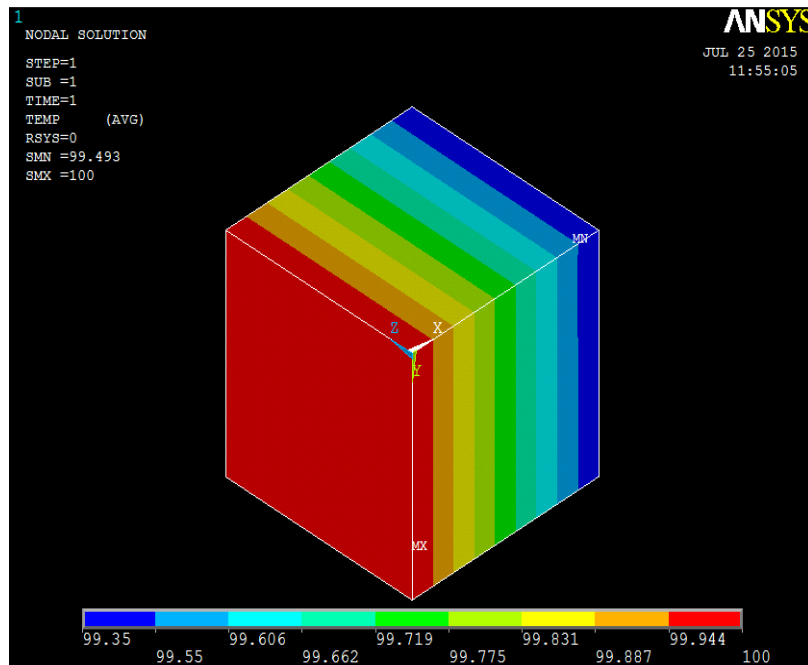
(c)



(d)



(e)



(f)

Fig. 5.4 Temperature profiles obtained for epoxy filled with (a) 1.41vol% TWD
(b) 3.4 vol% TWD (c) 6.5 vol% TWD (d) 11.3 vol% TWD
(e) 17.95 vol% TWD (f) 26.8 vol% TWD

5.2. Effective Thermal Conductivity: Comparison among results obtained from numerical and other existing theoretical correlations

From the temperature profiles, the value of effective thermal conductivity of each composite sample is estimated on the basis of one dimensional heat conduction equation. As expected, the value of k_{eff} comes out to be different for composites with different TWD content. These values obtained from FEM simulation for composites with different filler concentrations ranging from 0 to 26.8 vol% are presented in Table 5.1. It also presents the corresponding k_{eff} values calculated by using various existing theoretical correlations such as Rule of mixture, Maxwell's correlations [202] and Lewis and Nielsen's model [203] for composites having same filler concentrations. The comparison of these k_{eff} values obtained from numerical analysis and various theoretical correlations is illustrated graphically in Figure 5.5.

Table 5.1 Effective thermal conductivity of composites obtained from numerical analysis and various existing theoretical correlations

Volume fraction of filler (%)	Weight % of filler	Effective thermal conductivity(W/mK)			
		Rule of mixture	Maxwell's Correlation	Lewis and Nielsen's Model	FEM model
0	0	0.363	0.363	0.363	0.363
1.41	1.03	0.358	0.356	0.355	0.357
3.4	2.4	0.352	0.347	0.346	0.348
6.5	4.8	0.342	0.334	0.331	0.335
11.3	8.45	0.327	0.313	0.308	0.316
17.95	13.73	0.305	0.286	0.277	0.296
26.8	21	0.276	0.252	0.238	0.277

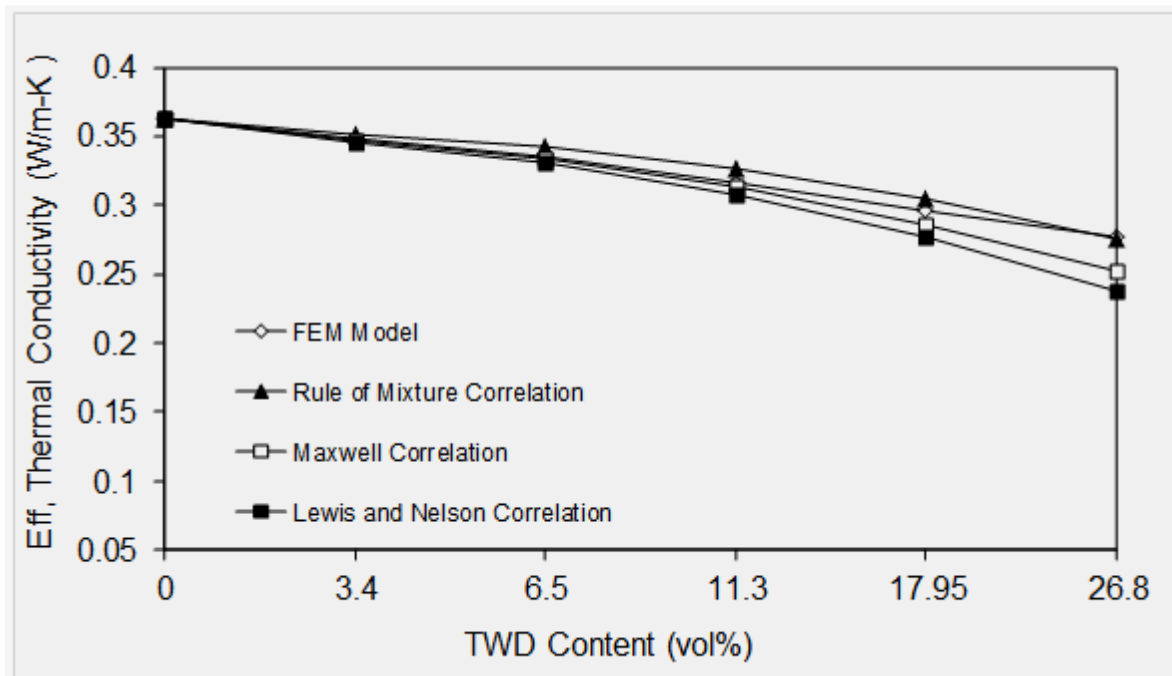


Fig. 5.5 Comparison of k_{eff} values obtained from numerical results and existing theoretical correlations

It is seen from Figure 5.5 that the values of effective thermal conductivity of composites obtained from different theoretical correlations are in good agreement with the values obtained from FEM model. With the increase in TWD content, a monotonic decrement in the value of k_{eff} is noticed irrespective of the method or model employed for the purpose.

5.3. Development of a theoretical model for estimation of effective thermal conductivity of a particulate-polymer composite system:

Heat transfer model for the present element

A 3-D schematic view of a particulate filled composite cube is shown in Figure 5.6 and a single element is taken out of it for further study of the heat transfer behavior as shown in Figure 5.7. The element consists of a small cube with a single particle in the center of it. The theoretical analysis of heat transfer in such a component is based on the following assumptions:

- Both the matrix and filler are locally homogeneous and isotropic.
- The composite lamina is free from voids.

- c) The temperature distribution along the direction of heat flow is linear.
- d) The filler-matrix interfacial thermal contact resistance is negligible.
- e) Density of matrix and filler will remain constant throughout the process of fabrication

On the basis of law of minimal thermal resistance and equal law of the specific equivalent thermal conductivity, when only one mode of heat transfer is considered i.e heat conduction, and specific equivalent thermal resistance of single element of the composite is considered equal to the total thermal resistance of the composite, then the equivalent thermal conductivity of that single element is considered equal to the total thermal conductivity of the composite.

Nomenclature:

H	➤	Side of the cube
r	➤	Radius of the filler
k_p	➤	Thermal conductivity of polymer matrix
k_f	➤	Thermal conductivity of filler phase
Q	➤	Heat quantity
Q_p, Q_f	➤	Heat quantity through the polymer matrix and filler respectively.
h_1	➤	$(H-2r) / 2$
S	➤	Total area of cross-section (heat transfer area)
S_p, S_f	➤	Cross-sectional area of the matrix and filler respectively
V_p, V_f, V_c	➤	Volume of matrix, filler and composite respectively
D_p, D_f, D_c	➤	Density of matrix phase, filler phase and composite respectively
R_1, R_2, R_3	➤	Heat resistance of part I, II and III respectively
R	➤	Total resistance of the element under study
ϕ	➤	Volume fraction of filler in the matrix
T	➤	Temperature
k_{eff}	➤	Effective thermal conductivity of element under study

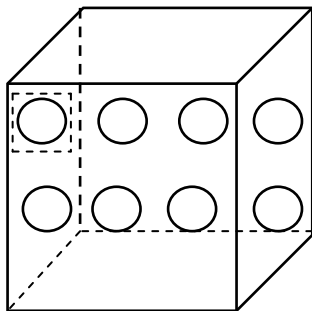


Fig. 5.6 3-D view of particulate filled composite cube

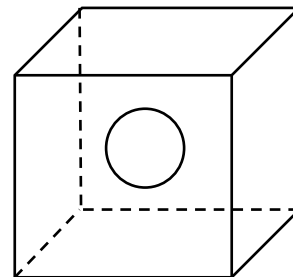


Fig. 5.7 3-D view of element under study

Figure 5.8 shows the front view of an element under study having side length H and a single spherical particle of radius r at the center. The filler material in the present work i.e. teak wood dust is nearly spherical in shape and hence for convenience, only the case of spherical inclusions is considered in the matrix. The element is divided into three parts, Part I, Part II and Part III having thermal conductivities k_1 , k_2 and k_3 respectively. S_p and S_f are the cross-sectional areas of the resin matrix and the filler, Q_p and Q_f are the heat quantities through the areas of the matrix and filler respectively. If we suppose that a composite is made up of a number of cubical elements and each element contains only a spherical particle at its center, then the total heat flow through the composite can be found out by considering the heat flow through a single element and then by integrating it for the entire composite system.

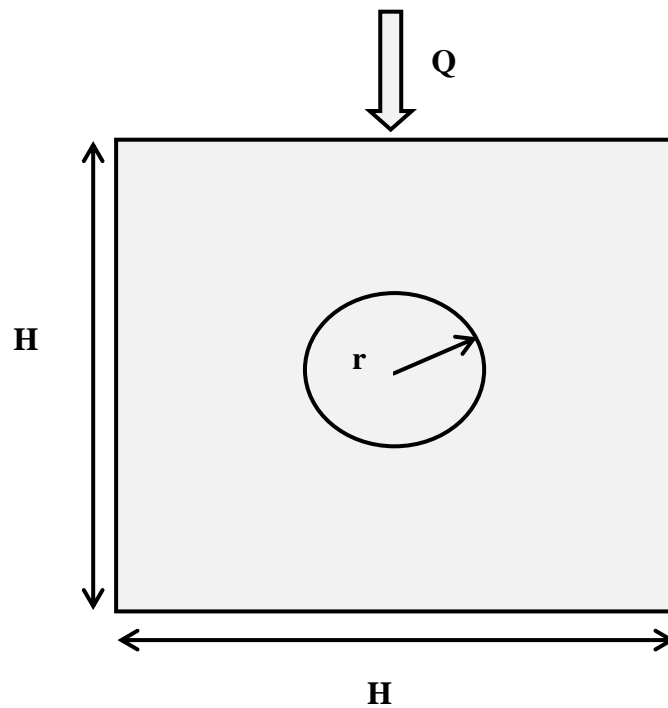


Fig.5.8 A heat transfer model for particulate-filled polymer composite

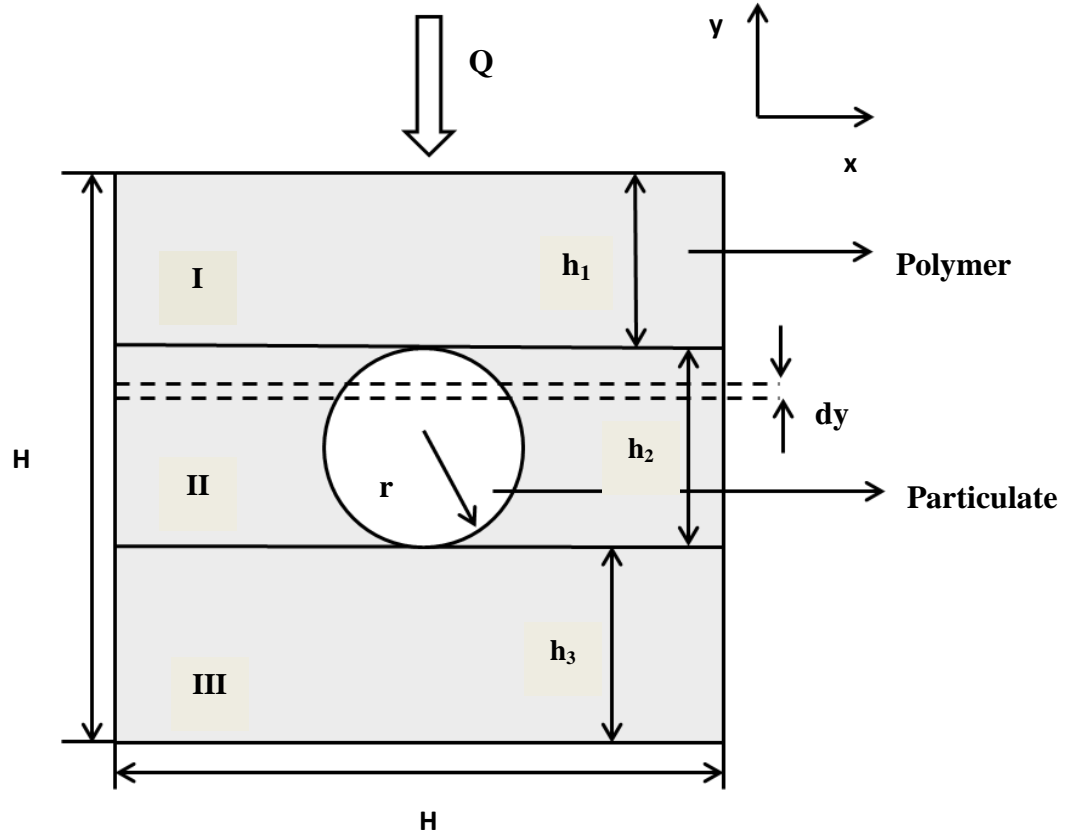


Fig.5.9 A series model of heat transfer in particulate-filled composite

The element is divided into a polymer phase and filler phase. The heat quantity Q flows from top to the bottom. The heat transfer across the particulate filled polymer composite occurs by two mechanisms: (i) solid thermal conduction and (ii) heat conduction on the surface between neighboring particles. The heat transfer is made mainly in the first way. The temperature changes more quickly inside the spherical particulate than outside the particulate. Polymer composite works usually under lower temperature conditions where the proportion of the thermal radiation in the total heat transfer is very small; hence the thermal radiation effect is neglected.

Part I and III:

$$k_1 = k_3 = k_p \quad (5.1)$$

Part II: Taking a thin piece with thickness of dy (as shown in Figure 5.9), according to Fourier's theorem, k_2 is given by:

$$k_2 = \frac{Q_p + Q_f}{\left(\frac{dT}{dy}\right)S} = \frac{k_p S_p}{S} + \frac{k_f S_f}{S} \quad (5.2)$$

Where, T is the temperature, S is the area of whole cross-section. k_p , k_f are the thermal conductivities of polymer matrix phase and micro-sphere phase. S_p and S_f are the cross-sectional areas of the polymer matrix and particulate. Q_p and Q_f are the heat quantities through the polymer matrix and particulate filler respectively. As the temperature distribution is linear, the average thermal conductivity of each section may first be obtained:

Part I and III:

$$k_1 = \int_{h_1} k_1 \left(\frac{dy}{h_1} \right) = k_p \quad (5.3)$$

Part II:

$$k_2 = \int_{h_2} k_2 \frac{dy}{h_2} = \int_{h_2} \left(\frac{k_p S_p}{S} + \frac{k_f S_f}{S} \right) \frac{dy}{h_2} = \frac{1}{h_2 S} (k_p S_p h_2 + k_f S_f h_2) = \frac{1}{h_2 S} (k_p V_p + k_f V_f) \quad (5.4)$$

According to the series theorem of heat resistance, the effective thermal conductivity of composite, k_{eff} is given by:

$$k_{eff} = \frac{H}{RS} = \frac{H}{(R_1 + R_2 + R_3 + R_{int})S} \quad R_1 = R_3, \quad R_{int} = 0 \quad (5.5)$$

Where, R_1 and R_2 are the heat resistance of part I and part II, R_3 is the heat resistance of part III, R is the total resistance of the element and R_{int} is the thermal contact resistance at the interface.

For particulate filled composite we have,

$$V_p D_p + V_f D_f = V_c D_c \quad (5.6)$$

Where, V_p and V_f are the volumes of polymer matrix phase and particulate phase respectively and V_c is the volume of the composite. Similarly, D_p and D_f are the densities of the polymer matrix phase and particulate filler phase respectively and D_c is the density of the composite as a whole.

The thermal resistance,

$$R = \frac{H}{kS} \quad (5.7)$$

$$R_1 = R_3 = \frac{h_1}{k_1 S} = \frac{h_1}{k_p S} h_1 = \frac{H - 2r}{2} \quad (5.8)$$

$$R_2 = \frac{h_2}{k_2 S} = \frac{h_2}{\left(\frac{k_p V_p + k_f V_f}{h_2 S} \right) S} = \frac{h_2^2}{(k_p V_p + k_f V_f)} = \frac{4r^2}{(k_p V_p + k_f V_f)} \quad , \quad h_2 = 2r \quad (5.9)$$

Where $2r$ is the particle diameter

The volume fraction of filler is given by

$$\phi = \frac{4\pi r^3}{3H^3} \quad (5.10)$$

$$H = r \left(\frac{4\pi}{3\phi} \right)^{\frac{1}{3}} \quad (5.11)$$

H is the length of the side of the cube.

Substituting the values of R_1 , R_2 and R_3 in the term of h_1 and h_2 in equation (5.5) we get,

$$k_{eff} = \frac{H}{\left(\frac{2h_1}{k_p S} + \frac{h_2^2}{(k_p V_p + k_f V_f)} \right) S} = \frac{H}{\frac{2h_1}{k_p} + \frac{4r^2 S}{(k_p V_p + k_f V_f)}} \quad (5.12)$$

$$k_{eff} = \frac{H}{\frac{2(H - 2r)}{2k_p} + \frac{4r^2 S}{(k_p V_p + k_f V_f)}} = \frac{1}{\frac{1}{k_p} \left(1 - \left(\frac{6\phi}{\pi} \right)^{\frac{1}{3}} \right) + \frac{4r^2 H}{(k_p V_p + k_f V_f)}} \quad , \quad S = H^2 \quad (5.13)$$

$$k_{eff} = \frac{1}{\frac{1}{k} \left(1 - \left(\frac{6\phi}{\pi} \right)^{\frac{1}{3}} \right) + \frac{2}{\frac{1}{2r^2 H} (k_p V_p + k_f V_f)}} \quad (5.14)$$

Substituting the value of V_p from equation (5.6), we get the final expression for effective thermal conductivity as:

$$k_{eff} = \frac{1}{\frac{1}{k_p} \left(1 - \left(\frac{6\phi}{\pi} \right)^{\frac{1}{3}} \right) + \frac{2}{\left\{ k_p \left(\frac{4\pi}{3\phi} \right)^{\frac{1}{3}} + \pi \left(\frac{2\phi}{9\pi} \right)^{\frac{1}{3}} \left(k_f \frac{D_p}{D_f} - k_p \right) \right\}}} \quad (5.15)$$

Here, k_p and k_f are the respective heat conductivities of the polymer and the particulate filler phase, D_p and D_f are the effective densities of the polymer and the filler phase respectively, ϕ is the volume fraction of the filler. The correlation given in the equation 5.15 can thus be used to estimate k_{eff} for such particulate filled composites with different filler content.

But since heat transfer within a filled polymer actually involves complex mechanisms, a simplified theoretical model for such a process may appear inadequate unless its assessment against experimental results is made. So for the validation of the proposed correlation i.e. Eqn. 5.15, thermal conductivity measurement tests on the composites are conducted using UnithermTM 2022 model under controlled laboratory conditions.

The measured values of effective thermal conductivity of epoxy-TWD composites for different filler content (1.41, 3.4, 6.5, 11.3, 17.95 and 26.8 vol %) are presented in Table 5.2. These values are compared with the theoretically predicted ones and the comparison is graphically illustrated in Figure 5.10.

Table 5.2 Effective thermal conductivities of composites: Theoretical and measured values

Volume fraction of filler (%)	Weight % of filler	Effective thermal conductivity(W/mK)					
		Rule of mixture	Maxwell	Lewis and Nielsen	Proposed model	Measured values	FEM model
0	0	0.363	0.363	0.363	0.363	0.363	0.363
1.41	1.03	0.358	0.356	0.355	0.359	0.333	0.357
3.4	2.4	0.352	0.347	0.346	0.352	0.320	0.348
6.5	4.8	0.342	0.334	0.331	0.342	0.290	0.335
11.3	8.45	0.327	0.313	0.308	0.326	0.240	0.3159
17.95	13.73	0.305	0.286	0.277	0.304	0.187	0.296
26.8	21	0.276	0.252	0.238	0.276	0.092	0.277

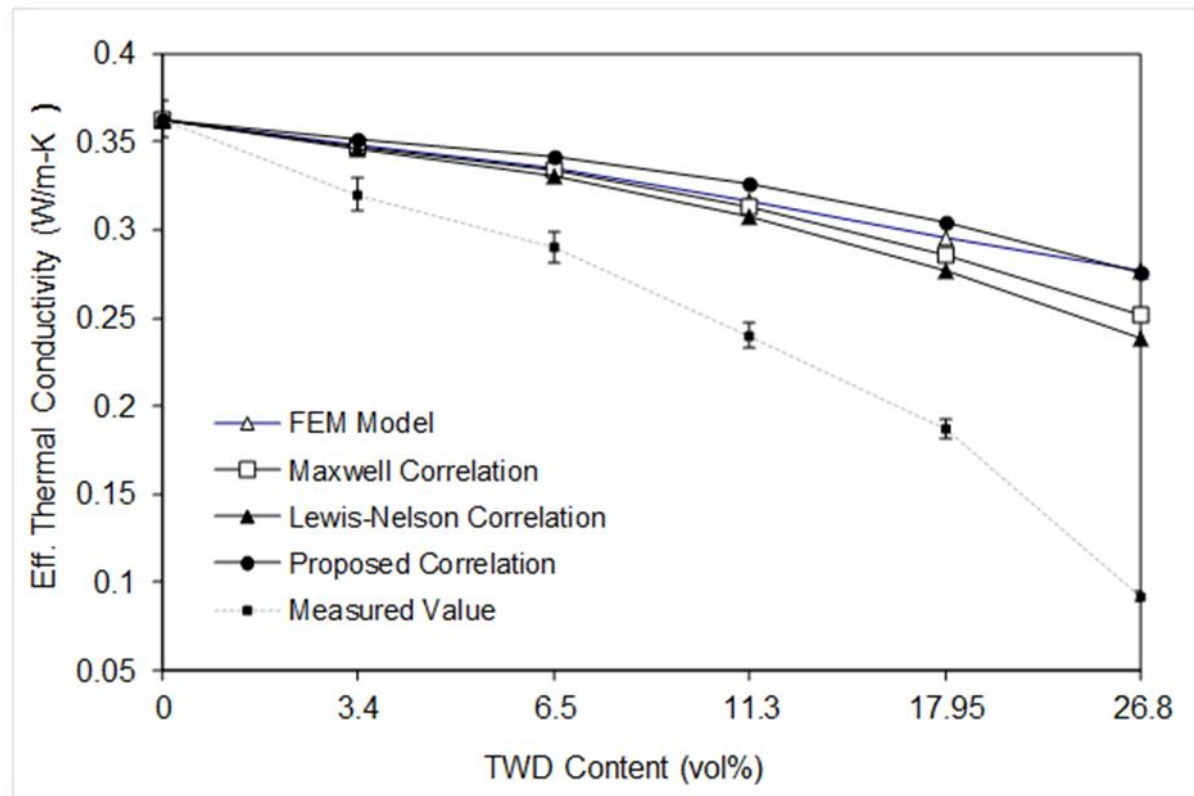


Fig.5.10 Comparison of k_{eff} values obtained from various methods

Although Figure 5.5 shows good agreement among the values, in regard to k_{eff} obtained from different theoretical models and numerical method, large discrepancies are noted between these values and the experimentally measured ones (Figure 5.10).

It is observed that the experimentally measured k_{eff} of the composites are significantly lower than all the theoretically predicted values irrespective of the filler concentration. This can partly be attributed to the presence of voids and pores within the composite body but the more possible reason is the thermal resistance at the filler-matrix interface. Polymers usually have hydrophobic surfaces therefore fillers with polar surfaces such as TWD would lead to poor interfacial bonding to the polymer further leading to high thermal contact resistance at the interfacial region. It is believed that the thermal contact resistance at the interface (R_{int}) plays a dominant role in significantly hindering the heat conduction capability of the composite system.

In the present research, the proposed correlation (Eqn. 5.15) does not consider the effect of thermal contact resistance at the interfacial region and hence overestimates the value of k_{eff} for composites with different filler concentrations. It is further noticed

that incidentally the difference between the predicted (Eqn. 5.15) and measured values of k_{eff} for any particular filler volume fraction happens to be proportional to the ratio of total filler-matrix interfacial area (A_{int}) to the volume of the composite (V). Therefore a new term $\psi * \left(\frac{A_{int}}{V} \right)$ is introduced to the correlation (Eqn. 5.15) to improve the accuracy of theoretical prediction. Hence, ψ is the proportionality constant and the new term in the modified correlation (Eqn. 5.16) takes into account the reduction in effective thermal conductivity of the composite due to interfacial thermal contact resistance.

$$k_{eff} = \frac{1}{\frac{1}{k_p} \left(1 - \left(\frac{6\phi}{\pi} \right)^{\frac{1}{3}} \right) + \frac{2}{\left\{ k_p \left(\frac{4\pi}{3\phi} \right)^{\frac{1}{3}} + \pi \left(\frac{2\phi}{9\pi} \right)^{\frac{1}{3}} \left(k_f \frac{D_p}{D_f} - k_p \right) \right\}}} - \psi \left(\frac{A_{int}}{V} \right) \quad (5.16)$$

From the above correlation (Eqn. 5.16), the effective thermal conductivities of the composites under study are estimated for different filler concentrations. In order to validate the correlation and to attain a close approximation with the experimental results a suitable value for ψ as 11×10^{-6} is chosen. Table 5.3 presents the values of k_{eff} obtained from different theoretical models including the modified correlation proposed in this work. These values are compared with the experimentally measured values of k_{eff} for epoxy-TWD composites of different compositions and the comparison is graphically illustrated in Figure 5.11.

Table 5.3 Comparison of measured k_{eff} values with those obtained from various methods

TWD content (vol%)	A_{int}/V (m^{-1})	Effective thermal conductivity(W/mK)						
		Rule of mixture	Maxwell	Lewis and Nelson	FEM model	Proposed correlation	Proposed correlation (modified)	Measured values
0	0	0.363	0.363	0.363	0.363	0.363	0.363	0.363
1.41	846	0.358	0.356	0.355	0.357	0.359	0.350	0.333
3.4	2040	0.352	0.347	0.346	0.348	0.352	0.330	0.320
6.5	3900	0.342	0.334	0.331	0.335	0.342	0.299	0.290
11.3	6780	0.327	0.313	0.308	0.316	0.326	0.251	0.240
17.9	10700	0.305	0.286	0.277	0.296	0.304	0.184	0.188
26.8	16080	0.276	0.252	0.238	0.277	0.276	0.099	0.092

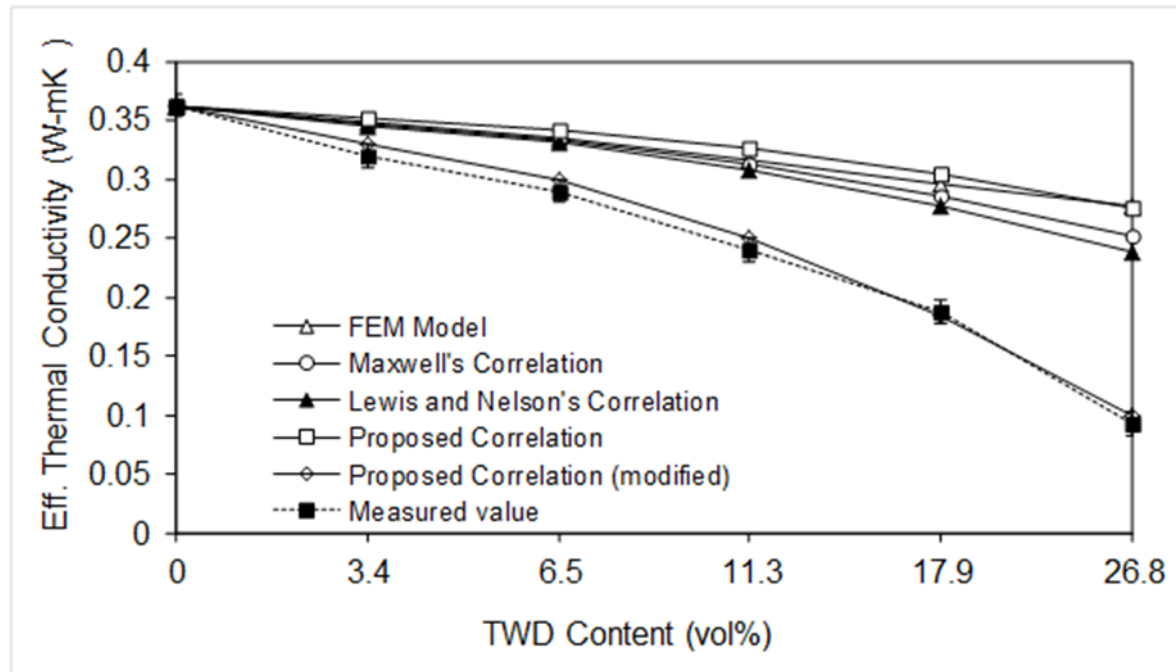


Fig. 5.11 Comparison of measured k_{eff} values with those obtained from various methods

In order to further validate the proposed correlation, the thermal conductivities of SET 2 composites are measured experimentally using the same UnithermTM 2022 conductivity tester. The measured values are compared with the theoretically estimated values (Eqn. 5.16) and the percentage error associated with each predicted values is presented in Table 5.4. The comparison graph showing a reasonably good agreement between the theoretical and experimental values is illustrated in Figure 5.12.

Table 5.4 Comparison of theoretical and experimental k_{eff} values of Epoxy-TWD composite

Composite Designation	TWD Content		Effective Thermal Conductivity (W/mK)		Error percentage (%)
	vol %	wt. %	Measured Value	Proposed Correlation (modified)	
ET0	0	0	0.363	0.363	0
ET5	5	3.69	0.309	0.314	1.618
ET10	10	7.47	0.253	0.264	4.348
ET15	15	11.37	0.202	0.214	5.941
ET20	20	15.40	0.148	0.165	11.486
ET25	25	20.30	0.098	0.116	18.367
ET30	30	23.76	0.058	0.066	19.846

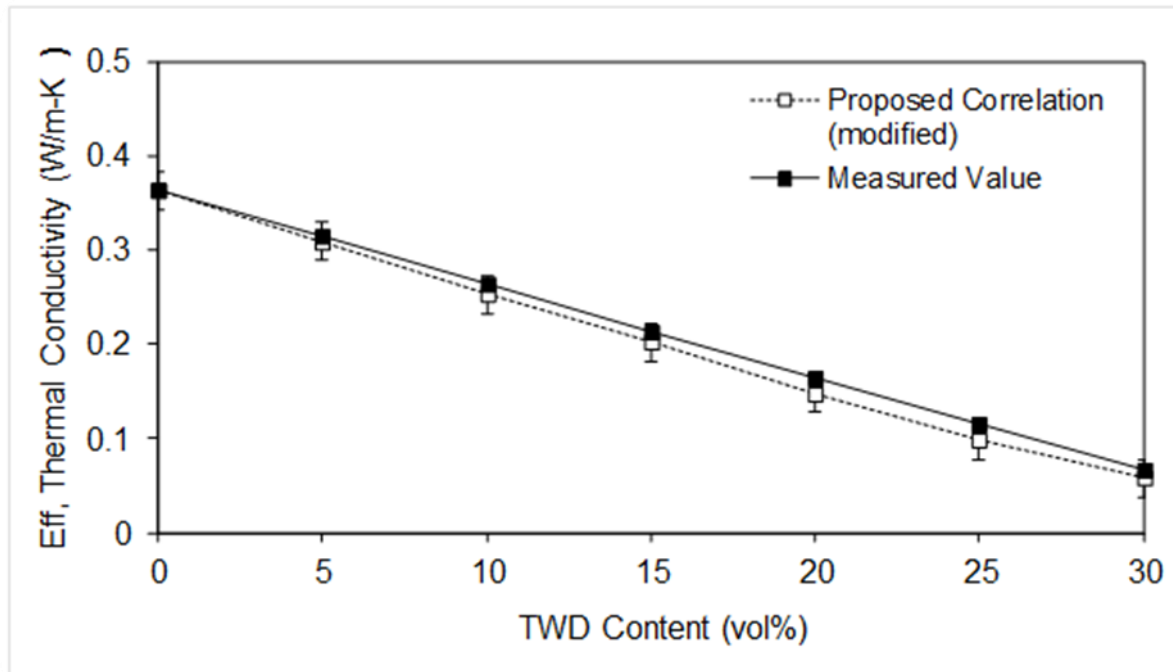


Fig. 5.12 Comparison of theoretical and experimental k_{eff} values of epoxy-TWD composite

It is evident from Table 5.4 and Figure 5.12 that the predicted values of effective thermal conductivities based on the proposed correlation with $\psi = 11 \times 10^{-6}$ are in close approximation with the measured values for epoxy-TWD composites with a wider range of filler concentrations (0-30 vol%). It may thus be stated that Eqn. 5.16 can be used as a predictive equation for other particulate-polymer composite systems by suitably tuning the value of ψ .

It is seen that incorporation of micro-sized TWD particles into epoxy appreciably enhances its heat insulation capability. Experimental results suggest that by adding 30 vol% of TWD, the thermal conductivity of epoxy can be reduced by about 84% (from 0.363 to 0.058 W/mK). It also becomes clear that this reduction is attributed not only to the relatively lower conductivity of TWD but also to the thermal contact resistance offered by the interfacial region of filler-matrix combination.

Filler-matrix interfacial area within a composite increases either by increasing the number of filler particles or by increasing the particle size. In both the cases the effective thermal conductivity, which is now known to be a function of interfacial area per unit volume is expected to vary. The variation of k_{eff} with increase in particle size

(radius of TWD particle) for different filler concentrations is thus studied (Figure 5.13).

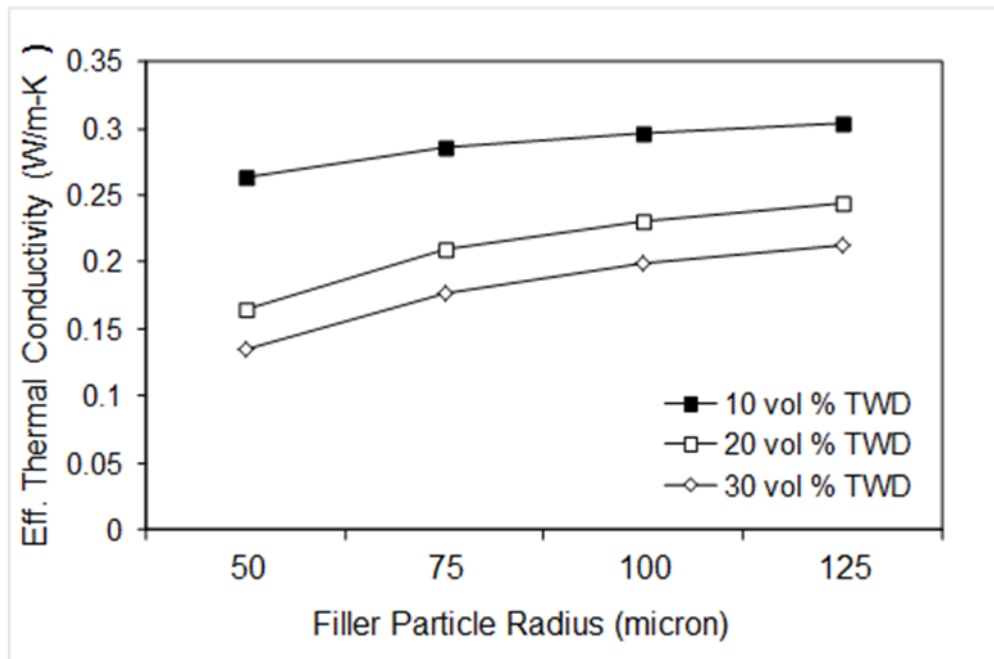


Fig. 5.13 Variation of k_{eff} as a function of filler size

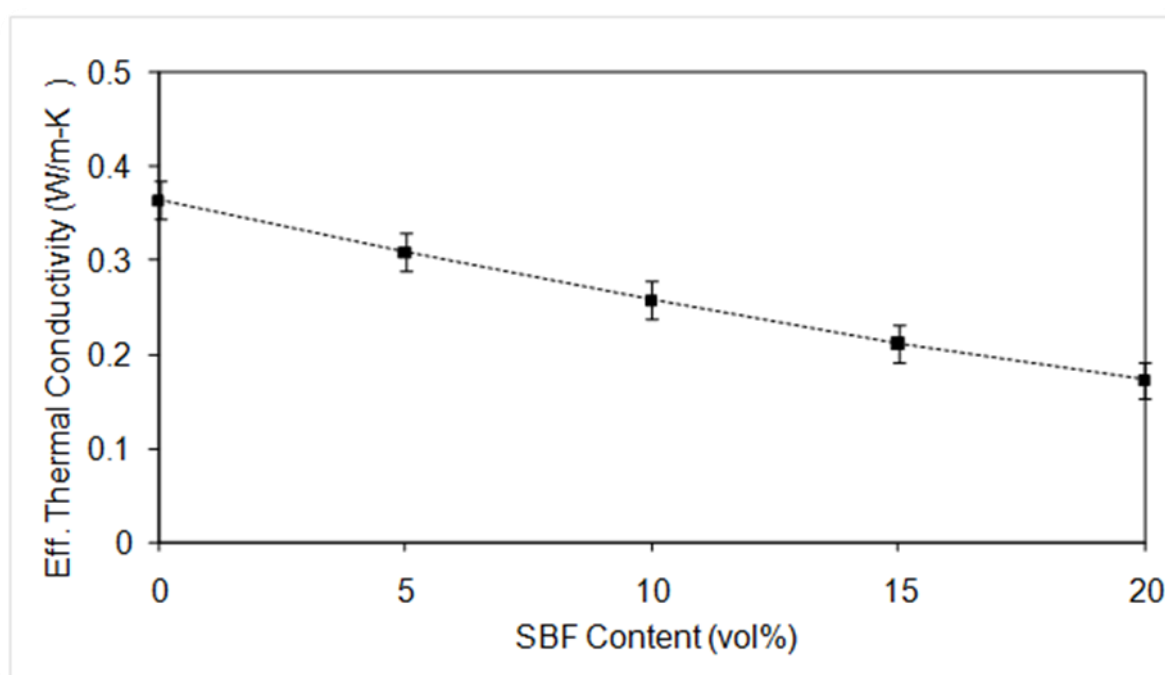
It is seen that as the filler radius increases from 50 to 125 micron, the effective thermal conductivity of the epoxy TWD composites improves irrespective of the filler content.

5.4. Effects of Banana Fiber Reinforcement on Effective Thermal Conductivity of Epoxy: k_{eff} of Epoxy-SBF Composites

The thermal conductivities of SET 3 samples i.e. epoxy-SBF composites are measured experimentally using the same UnithermTM 2022 conductivity tester. It is seen that the incorporation of short banana fiber into epoxy appreciably enhances its heat insulation capability. Table 5.5 presents the experimental results in regard to the k_{eff} values of these composites reinforced with different amounts of short banana fibers. With 5 vol% of SBF, the conductivity value drops from 0.363 W/mK to about 0.308 W/mK. Similarly, with addition of 20 vol% of SBF, k_{eff} of epoxy reduces by about 52%. The variation of k_{eff} of epoxy composites with volume fraction of SBF is graphically shown in Figure 5.14.

Table 5.5 Effective thermal conductivity as a function of SBF content

Composite Designation	SBF Content		Effective Thermal Conductivity (W/mK)
	vol %	wt. %	
EB0	0	0	0.363
EB5	5	0.95	0.308
EB10	10	1.98	0.257
EB15	15	3.10	0.211
EB20	20	4.35	0.172

**Fig. 5.14** Variation of k_{eff} with fiber content**Table 5.6** Effective thermal conductivity of hybrid composites (Epoxy-TWD-SBF)

Composite Designation	TWD Content (vol %)	SBF Content (vol %)	Effective Thermal Conductivity (W-mk) (measured)
ETB1	10	0	0.253
ETB2	10	5	0.232
ETB3	10	10	0.217
ETB4	20	0	0.148
ETB5	20	5	0.129
ETB6	20	10	0.103

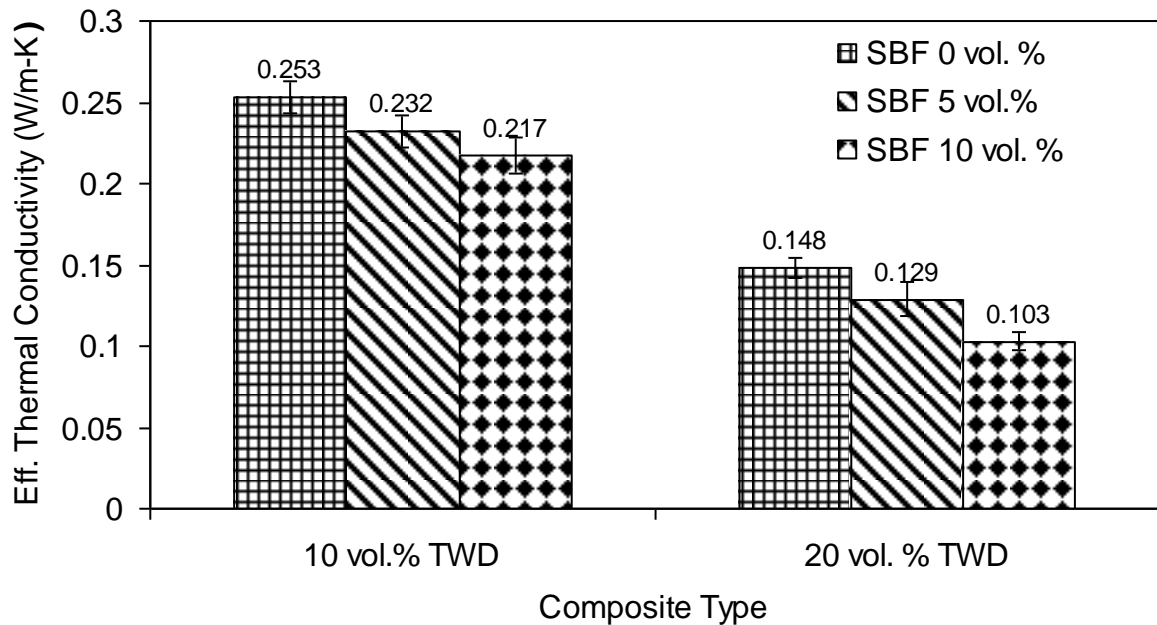


Fig. 5.15 Effective thermal conductivity of hybrid composites (Epoxy-TWD-SBF)

5.5. Effects of Hybrid Reinforcement on Effective Thermal Conductivity of Epoxy: k_{eff} of Epoxy-TWD-SBF Hybrid Composites

For multi filler composites, the effective thermal conductivity values of epoxy reinforced with both TWD and SBF (SET 4 composites) in different proportions are shown in Table 5.6. It has already been seen that, the incorporation of either of the TWD or SBF fillers in the epoxy enhances the heat insulation capability of the composite. But when certain amount of TWD is replaced by equal volume of SBF, the k_{eff} value marginally increases by an appreciable amount. This is due to the obvious reason that the intrinsic conductivity of SBF is more than that of epoxy-TWD.

5.6. Glass Transition Temperature (T_g)

The glass transition temperature (T_g) of the composite samples are measured with a Perkin Elmer DSC-7 thermal mechanical analyzer (TMA). Figure 5.16 presents the variation of T_g with TWD content. It is observed that the T_g of neat epoxy is about 98°C

and it gradually increases to 112°C with an increase in TWD content as shown in Table 5.7. A maximum increase of 14°C in T_g is obtained as the TWD content in the composite is increased from 0 to 40 vol%.

Table 5.7 variation of T_g and CTE of polymer composite with respect to filler content

TWD content (vol%)	T_g (°C)	CTE (ppm/°C)
0	98.0	66.0
5	98.7	65.9
10	100.3	65.6
15	102.4	65.2
20	105.0	64.6
25	107.5	63.9
30	109.0	63.5
35	110.4	63.1
40	112.0	62.6

The effect of filler addition in polymer composites on glass transition behavior has been investigated in the past for different particulate filled polymer composites [222,223] and it has been seen that addition of micro-sized fillers usually improves the T_g of composites [223]. In the present study, a shift of T_g towards a higher value can be attributed to the filler-matrix interaction, which might be restricting the mobility of the epoxy chain.

5.7. Coefficient of Thermal Expansion (CTE)

The CTE of the TWD filled epoxy composites are measured with thermal mechanical analyzer (TMA). Figure 5.17 shows that the CTE of the composite samples reduces with the increase in TWD content. The CTE of neat epoxy is about 66 ppm/°C and it gradually reduces to 62.6 ppm/°C with increase in filler content. A maximum decrease of about 5.15 % in CTE is obtained with 40 vol% of TWD in the composite as shown in Table 5.7.

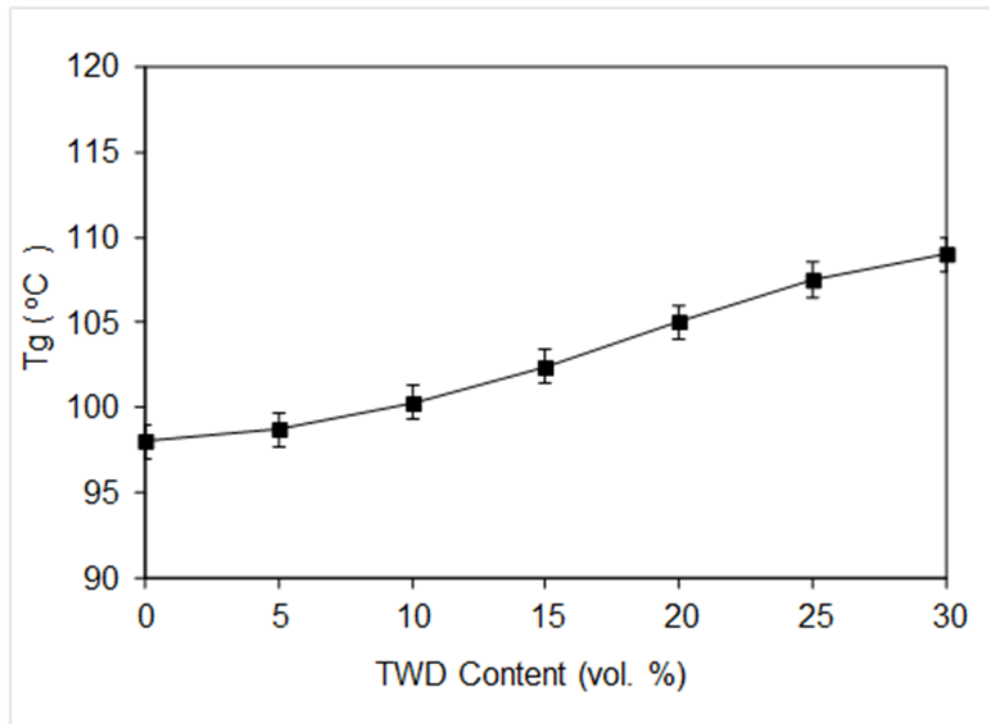


Fig. 5.16 Variation of T_g of epoxy-TWD composites with filler content

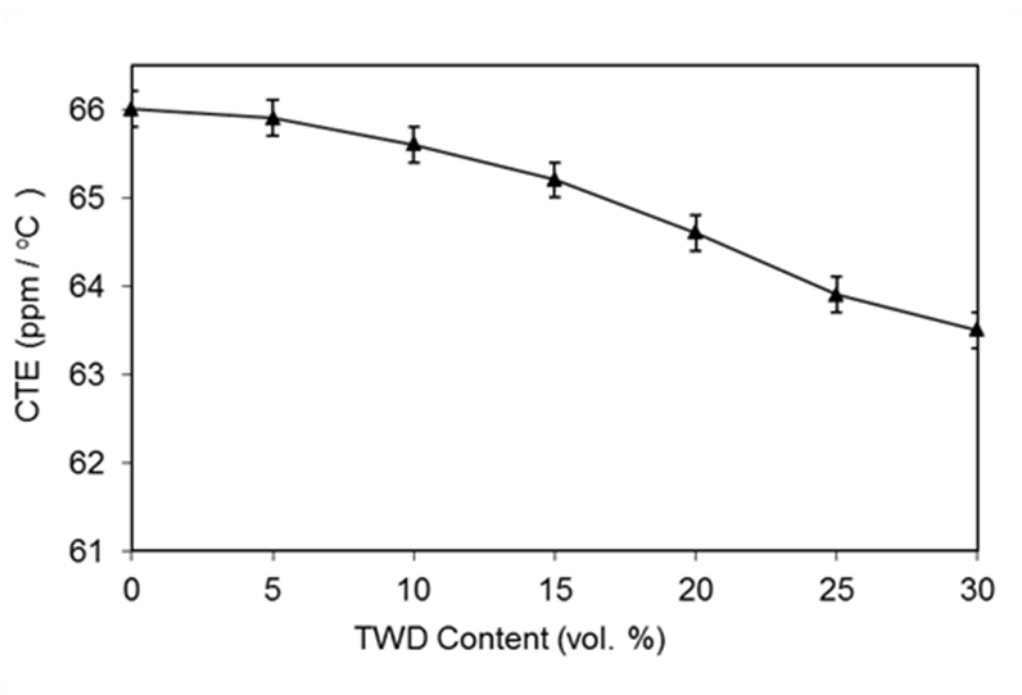


Fig. 5.17 Variation of CTE of epoxy-TWD composites with filler content

The constraint of mobility of the epoxy chain due to the interaction of TWD and epoxy are responsible for the reduction of CTE. Hence, the thermal stability of epoxy improves with addition of TWD particles.

Chapter Summary

This chapter has provided:

- The results of the numerical analysis and experiments conducted to evaluate the effective thermal conductivity of the polymer composites under study.
- The validation of proposed theoretical model through experimental results.
- The effects of TWD and SBF on the thermal insulation capability of epoxy.
- The effects of TWD on the glass transition temperature and coefficient of thermal expansion of composites.

The next chapter presents a summary of the research findings and the specific conclusions drawn from this work. Some potential applications of the fabricated composites have also been recommended. It also suggests ideas and directions for future research in the related field.

CHAPTER- 6

Summary and Conclusions

Chapter 6**SUMMARY AND CONCLUSIONS**

The research reported in this thesis broadly consists of three parts:

- The first part has provided the description of the materials used and the details of the experiments that are carried out during this research. It also presents the test results in regard to the physical, mechanical and micro-structural characteristics of the epoxy filled with micro-sized teak wood dust and/or short banana fibers.
- The second part is about the development of a theoretical heat conduction model based on which a mathematical correlation has been proposed for estimation of effective thermal conductivity of polymer composites with uniformly distributed micro-sized particulate fillers. In this part, the correlation is validated for teak wood dust (TWD) filled polymers through numerical analysis and experimentation.
- The last part has presented the experimental results related to the effective thermal conductivity of hybrid composites filled with teak wood dust and short banana fibers (SBF). This part has also reported on these epoxy composites in regard to their other thermal characteristics such as glass transition temperature (T_g) and coefficient of thermal expansion (CTE).

6.1 Summary of Research Findings

The performance of any new composite product is often judged by its response under different physical, mechanical and thermal conditions as it becomes essential for selecting the material of proper composition for any given application. Therefore, in

the present work, a wealth of property data has been generated for a series of epoxy-TWD, epoxy-SBF and epoxy-TWD-SBF hybrid composites by fabricating them in the laboratory and by conducting various physical, mechanical and thermal tests on them under controlled laboratory conditions. An attempt has been made to develop a semi-empirical approach so as to include the effect of thermal contact resistance at the filler-matrix interfacial region and subsequently a mathematical correlation is proposed to estimate the effective thermal conductivity of particulate filled composites.

It is found that by incorporating micro-sized TWD particulates into epoxy resin, its effects, as expected are achieved in the form of modified physical, mechanical and thermal properties. Due to the presence of TWD micro-fillers, changes in their heat conduction behavior are seen. When TWD is added in epoxy matrix, the effective thermal conductivity of the composite is reduced as wood dust is thermally insulative in nature. Additional reinforcement of short banana fibers in epoxy-TWD composite system further reduces the effective conductivity while enhancing the strength properties. Effects of TWD and SBF on the glass transition temperature and coefficient of thermal expansion of epoxy resin are also found to be substantial.

6.2 Conclusions

This analytical and experimental investigation on epoxy composites filled with TWD micro-particles and/or SBF has led to the following specific conclusions:

1. Successful fabrication of epoxy-TWD, epoxy-SBF and epoxy-TWD-SBF hybrid composites by hand layup technique is possible.
2. Incorporation of TWD particles and short banana fibers modifies the tensile, compressive and flexural strengths of the composites. It is noticed that with addition of TWD particles, the strength of epoxy is marginally decreased and this decrement is a function of the filler content. On the contrary, reinforcement

- of SBF enhances the tensile, compressive and flexural strengths of these epoxy based composite systems.
3. A theoretical correlation to estimate the effective thermal conductivity of particulate filled composites is proposed based on the one dimensional heat conduction model. It is seen that this serves as a good semi-empirical model for spherical inclusions and that the proposed correlation can therefore very well be used to estimate the effective thermal conductivity for particulate filled composites.
 4. It is propitious that the incorporation of TWD particles results in significant improvement in the thermal insulation capability of epoxy resin. It is found that with increase in the TWD content from 0 to 30 vol%, the value of effective thermal conductivity (k_{eff}) drops by about 84% for epoxy.
 5. Reinforcement of short banana fibers in epoxy also helps in improving the insulation characteristics of epoxy resin. With the incorporation of 20 vol% of SBF, the thermal conductivity of epoxy reduces from 0.363W/mK to 0.172W/mK (by about 52%). Hybrid composites comprising of both SBF and TWD micro-fillers also exhibit improved insulation capabilities as compared to neat epoxy.
 6. It is observed that the T_g of epoxy (98°C) gradually increases to 112°C as the TWD content increases from 0 to 40 vol%. This increase in T_g can be attributed to strong interaction between the filler/fiber and the epoxy-matrix which restricts the mobility of the polymer chain. It is further seen that with the incorporation of TWD, the coefficient of thermal expansion (CTE) of epoxy is substantially lowered.
 7. With light weight, low thermal expansion coefficient and improved insulation capability, the teak wood dust and short banana fiber reinforced epoxy

composites can be used for applications such as insulation boards, food containers, thermo flasks, refrigeration industry, building materials, interiors of air crafts and automobiles etc.

6.3 Scope for Future Research

The present research work leaves a wide scope for future investigators to explore many other aspects of such particulate filled composites. Some recommendations for future research include:

- Possible use of organic fillers other than teak wood dust and polymeric resins other than epoxy in the development of new composites.
- Exploring the possibility of using natural fibers other than banana fibers along with ceramic particulates to fabricate hybrid composites with improved functional properties.
- Cost analysis of these composites to assess their economic viability in domestic/industrial applications.

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APPENDICES

Appendix-A1

List of publications from this research work

International Journals

1. **Mimosha Dash**, Alok Agrawal and Alok Satapathy, *Effective thermal conductivity of epoxy composites filed with micro-sized teak wood dust particles: An analytical and experimental investigation*, **Journal of Reinforced Plastics and Composites** (Under Review)

International Conferences

1. **Mimosha Dash** and Alok Satapathy, *A study on effective thermal conductivity of fiber and particulate reinforced polymer composites*, Proceedings of International Conference on Advancement in Polymeric Materials APM-2015, February, 2015, IISC Bangalore.
2. **Mimosha Dash** and Alok Satapathy, *Numerical and analytical modeling for heat transfer through particulate reinforced polymer composite*, Proceedings of International Seminar on Renewable Energy and Sustainable Development, June, 2015, Royal University of Bhutan, Thimphu.

Appendix- A2

Brief Bio-data of the Author

The author, **Mimosha Dash**, born on 01-06-1992 graduated in Mechanical Engineering from Biju Patnaik University of Technology (BPUT) in the year 2013. She joined National Institute of Technology, Rourkela in the year 2013 as an M.Tech. Research Scholar in the Department of Mechanical Engineering. In the postgraduate research programme, she has worked in the area of *thermal characteristics of particulate/fiber filled polymer composites*. She has communicated 01 research paper to an International journal and has presented 02 research papers in the area of polymer composites at various international conferences

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Thermal conductivity of particulate reinforced epoxy composites: an analytical and experimental investigation

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Manuscript Type:	Original Manuscript
Date Submitted by the Author:	n/a
Complete List of Authors:	Dash, Mimosh; N.I.T. Rourkela, Mechanical Engineering AGRAWAL, ALOK; NATIONAL INSTITUTE OF TECHNOLOGY, ROURKELA, MECHANICAL ENGINEERING DEPARTMENT Satapathy, Alok; N.I.T. Rourkela, Mechanical Engineering
Keywords:	Polymer matrix composites, Epoxy, Teak wood dust, Effective thermal conductivity, Analytical modeling, Finite element method, Coefficient of thermal expansion, Glass transition temperature.
Abstract:	<p>This paper depicts the processing and characterization of a class of particulate reinforced polymer composites consisting of epoxy resin filled with teak wood dust (TWD) particulates. The effects of incorporation of TWD in modifying the physical and mechanical properties of epoxy composites are studied. Effective thermal conductivity values are also estimated numerically by finite element method using a commercially available finite-element package ANSYS. Further, a mathematical correlation for the effective thermal conductivity of such particulate-filled polymer composites is developed using the law of minimal thermal resistance and equal law of specific equivalent thermal conductivity. To validate this correlation, two sets of epoxy-based composites with micro-sized TWD are prepared by hand-lay-up technique. Thermal conductivities of these composites are measured using the Unitherm™ Model 2022 tester. These values are then compared with the values obtained from the proposed model and are found to be in fairly good agreement. This study shows that the incorporation of teak wood dust results in reduction of conductivity of epoxy resin and thereby improves its thermal insulation capability. With addition of 5 vol % of filler, the thermal conductivity of epoxy is found to decrease by about 15% and with about 30 vol % of filler addition, a 84% reduction in thermal conductivity of neat epoxy is achieved. Effects of fillers on other thermal properties like coefficient of thermal expansion, glass transition temperature of epoxy resin are also studied.</p>

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Thermal conductivity of particulate reinforced epoxy composites: an analytical and experimental investigation

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Abstract

This paper depicts the processing and characterization of a class of particulate reinforced polymer composites consisting of epoxy resin filled with teak wood dust (TWD) particulates. The effects of incorporation of TWD in modifying the physical and mechanical properties of epoxy composites are studied. Effective thermal conductivity values are also estimated numerically by finite element method using a commercially available finite-element package ANSYS. Further, a mathematical correlation for the effective thermal conductivity of such particulate-filled polymer composites is developed using the law of minimal thermal resistance and equal law of specific equivalent thermal conductivity. To validate this correlation, two sets of epoxy-based composites with micro-sized TWD are prepared by hand-lay-up technique. Thermal conductivities of these composites are measured using the Unitherm™ Model 2022 tester. These values are then compared with the values obtained from the proposed model and are found to be in fairly good agreement. This study shows that the incorporation of teak wood dust results in reduction of conductivity of epoxy resin and thereby improves its thermal insulation capability. With addition of 5 vol % of filler, the thermal conductivity of epoxy is found to decrease by about 15% and with about 30 vol % of filler addition, a 84% reduction in thermal conductivity of neat epoxy is achieved. Effects of fillers on other thermal properties like coefficient of thermal expansion, glass transition temperature of epoxy resin are also studied.

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Keywords

Polymer matrix composites, epoxy, teak wood dust, effective thermal conductivity, analytical modeling, finite element method, coefficient of thermal expansion, glass transition temperature.

Introduction

Insulation to retard the flow of heat is important for comfort and in some cases, for the survival of humans and animals. It also prevents damage to various articles by freezing or by high temperatures and lowers heating and cooling costs. The potential impact of increasing energy efficiency of engineering components and thereby reducing the greenhouse gas emission through better thermal insulation of engineering structures and buildings is enormous. To achieve this, the most obvious approach would be to apply thicker layers of readily available thermal insulation materials. But this can locally result in the use of undesirable thick constructions and related changes in building methods, not to mention the loss of valuable floor space and the higher costs. Instead of this option, novel types of affordable and processable thermal insulation materials with improved efficiencies are being considered [1]. Among the best thermal insulation materials available today are vacuum insulation panels [2] and aerogel based materials [3]. But in spite of their high insulation efficiencies, these materials suffer from processability and pricing issues that restrict their wide spread utilization [3, 4].

The conventional engineering materials are often unable to meet the requirement of these special properties like high strength, low density and low conductivity. The demand for structurally stable, cost effective and light-weight insulation materials is therefore increasing day by day. This is exactly why industrial and research efforts are directed towards the development of plastic foams and/or polymer matrix composites, eventually allowing the

production of a new class of affordable, highly efficient thermal insulation materials. At present, the widely used insulation materials in the industries are based either on glass fibers, which are derived from the silica sources or on other synthetic fibers derived from the petroleum based resources. Unfortunately, glass fiber based materials are known to have carcinogenic effects [5] and petroleum based materials are non-renewable. This opens up another option i.e. polymer composites based on natural fibers.

Considerable work has been reported in the past on the subject of heat conductivity in polymers by Hansen and Ho [6], Peng and Landel [7], Choy and Young [8], Tavman [9] etc. But most of these studies were confined to the thermal behaviour of neat polymers only and not to their composites. Reports are available in the existing literature on experimental as well as numerical and analytical studies on thermal conductivity of some filled polymer composites [10-14]. The fillers most frequently used are aluminum, copper, brass particles, short carbon fiber, carbon particles, graphite, aluminum nitrides and magnetite particles. Progelhof et al. [10] were the first to present an exhaustive overview on models and methods for predicting the thermal conductivity of composite systems. Many other theoretical and empirical models have also been proposed to predict the effective thermal conductivity of two-phase mixtures. Comprehensive review articles are available on the applicability of many of these models [15, 10]. Apart from analytical study, few reports on numerical approach to predict the effective thermal conductivity of composite materials were proposed by Kumultas and Tavman [16] and Nayak et al. [17].

Theory

Existing theoretical models for effective thermal conductivity of composites

It has been seen in the literature that addition of fillers to a polymer, changes its behavior, and a significant alteration in the effective thermal conductivity (k_{eff}) of the system has been observed. Therefore, it is very important to understand the heat transfer mechanism in

polymer composites. To predict the effective thermal conductivity of composite materials, several theoretical and empirical models have been proposed in the past. The two basic equations of thermal conduction used in composites are Rule of Mixture derived on the basis of series conduction and parallel conduction respectively [16].

Parallel conduction model:

$$k_{eff} = (1 - \phi_f)k_p + \phi_f k_f \tag{1}$$

Series conduction model:

$$\frac{1}{k_{eff}} = \frac{(1 - \phi_f)}{k_p} + \frac{\phi_f}{k_f} \tag{2}$$

Maxwell used the potential theory and obtained an “exact” solution for the conductivity of randomly distributed and non-interacting homogeneous spheres in a homogeneous continuous medium [18] which is expressed as,

$$\frac{k_{eff}}{k_p} = \left(\frac{k_f + 2k_p + 2\phi_f(k_f - k_p)}{k_f + 2k_p - \phi_f(k_f - k_p)} \right) \tag{3}$$

Bruggeman [19] derived the implicit equation for dilute suspension of sphere in a homogeneous medium and gave it an expression as follows,

$$1 - \phi_f = \frac{k_{eff} - k_f}{k_p - k_f} \left(\frac{k_p}{k_{eff}} \right)^{\frac{1}{3}} \tag{4}$$

Lewis and Nielsen [20] included the effect of the shape of the particles and its orientation for a two-phase system for calculating thermal conductivity and derived the following correlation.

$$k_{eff} = k_p \left(\frac{1 + AB\phi}{1 - B\phi\psi} \right) \tag{5}$$

where

$$B = \frac{\frac{k_f}{k_p} - 1}{\frac{k_f}{k_p} + A} \quad \text{and} \quad \psi = 1 + \left(\frac{1 - \phi_p}{\phi_p^2} \right) \phi$$

A and B are the constants and are having a specific values for a particular shape and size of filler particles.

The model proposed by Lewis and Nielsen [20] can better estimate the effective thermal conductivity of filled composites, while the Agari–Nagai equation can predict the effective thermal conductivity of composites with high-loading [21]. In a most recent work, Agrawal and Satapathy [22, 23] proposed models for single filler as well as for multi filler composites. In spite of so many existing theoretical models, heat transfer through a heterogeneous system like polymer composite has remained a challenging subject for the researchers as no single model predicts the effective thermal conductivity for all filler type, size, shape, concentrations and interface thermal resistance. Against this background, this work attempts to develop theoretical and numerical models for estimation of effective thermal conductivity of particulate filled polymer composites and to validate the models for an epoxy based composite system filled with micro-sized teak wood dust through experimentation.

Development of a theoretical model for estimation of effective thermal conductivity of a particulate-polymer composite system

Nomenclature:

H	➤	side of the cube
r	➤	radius of the particulate filler
k_p	➤	thermal conductivity of polymer matrix
k_f	➤	thermal conductivity of filler phase
Q	➤	heat quantity
Q_p, Q_f	➤	heat quantity through the matrix and filler respectively
h_l	➤	$(H-2r) / 2$
S	➤	total area of cross-section (heat transfer area)
S_p, S_f	➤	cross-sectional area of the matrix and filler respectively
V_p, V_f, V_c	➤	volume of matrix, filler and composite respectively
D_p, D_f, D_c	➤	density of matrix phase, filler phase and composite respectively
R_1, R_2, R_3	➤	heat resistance of part 1, 2 and 3 respectively
Φ_f	➤	volume fraction of filler in the matrix
T	➤	temperature

Heat transfer model for the present element. A 3-D schematic view of a particulate filled composite cube is shown in Figure 1 (a) and a single element is taken out of it for further study of the heat transfer behavior as shown in Figure 1 (b). The element consists of a small cube with a single particle in the center of it.

The theoretical analysis of heat transfer in such a component is based on the following assumptions:

- a) Both the matrix and filler are locally homogeneous and isotropic;
- b) The composite lamina is free from voids;
- c) The temperature distribution along the direction of heat flow is linear;
- d) The filler-matrix interfacial thermal contact resistance is negligible;
- e) Density of matrix and filler will remain constant throughout the process of fabrication.

On the basis of law of minimal thermal resistance and equal law of the specific equivalent thermal conductivity, when only one mode of heat transfer is considered i.e. heat conduction, and specific equivalent thermal resistance of single element of the composite is considered equal to the total thermal resistance of the composite, then the equivalent thermal conductivity of that single element is considered equal to the total thermal conductivity of the composite. Figure 2 shows the front view of an element under study having side length H and a single spherical particle of radius r at the center. The heat quantity Q flows from top to the bottom. The filler material in the present work i.e. teak wood dust is nearly spherical in shape and hence for convenience, only the case of spherical inclusions is considered in the matrix. The element is divided into three parts as shown in Figure 3 i.e. Part I, Part II and Part III having thermal conductivities k_1 , k_2 and k_3 respectively.

Part I and III:

$$k_1 = k_3 = k_p \quad (6)$$

Part II: Taking a thin piece with thickness of dy (as shown in Figure 3), according to

Fourier's theorem, k_2 is given by:

$$k_2 = \frac{Q_p + Q_f}{\left(\frac{dT}{dy}\right)S} = \frac{k_p S_p}{S} + \frac{k_f S_f}{S} \quad (7)$$

As the temperature distribution is considered linear, the average thermal conductivity of each section may first be obtained.

Part I and III:

$$k_1 = k_3 = \int_{h_1} \left(\frac{dy}{h_1} \right) = k_p \quad (8)$$

Part II:

$$k_2 = \int_{h_2} k_2 \frac{dy}{h_2} = \int_{h_2} \left(\frac{k_p S_p}{S} + \frac{k_f S_f}{S} \right) \frac{dy}{h_2} = \frac{1}{h_2 S} (k_p S_p h_2 + k_f S_f h_2) = \frac{1}{h_2 S} (k_p V_p + k_f V_f) \quad (9)$$

According to the series theorem of heat resistance, the effective thermal conductivity of composite, k_{eff} is given by:

$$k_{eff} = \frac{H}{RS} = \frac{H}{(R_1 + R_2 + R_3 + R_{int})S} \quad R_1 = R_3, \quad R_{int} = 0 \quad (10)$$

where, R_{int} is the thermal contact resistance at the interface.

As it is assumed that the density of matrix and filler will remain constant throughout the process of fabrication,

$$V_p D_p + V_f D_f = V_c D_c \quad (11)$$

Also, thermal resistance is given by,

$$R = \frac{H}{kS} \tag{12}$$

So for Part *I*, *II* and *III* thermal resistance can be given as,

$$R_1 = R_3 = \frac{h_1}{k_1 S} = \frac{h_1}{k_p S} \quad \text{where, } h_1 = \frac{H - 2r}{2} \tag{13}$$

$$R_2 = \frac{h_2}{k_2 S} = \frac{h_2}{\left(\frac{k_p V_p + k_f V_f}{h_2 S} \right) S} = \frac{h_2^2}{(k_p V_p + k_f V_f)} = \frac{4r^2}{(k_p V_p + k_f V_f)} \quad \text{where, } h_2 = 2r \tag{14}$$

Where $2r$ is the particle diameter

The volume fraction of filler is given by

$$\phi = \frac{4\pi r^3}{3H^3} \tag{15}$$

This may be rearranged and can be written as,

$$H = r \left(\frac{4\pi}{3\phi} \right)^{1/3} \tag{16}$$

Substituting the values of R_1 , R_2 and R_3 in terms of h_1 and h_2 in equation (10),

$$k_{eff} = \frac{H}{\left(\frac{2h_1}{k_p S} + \frac{h_2^2}{k_p V_p + k_f V_f} \right) S} = \frac{H}{\frac{2h_1}{k_p} + \frac{4r^2 S}{(k_p V_p + k_f V_f)}} \tag{17}$$

Putting the value of h_1 from equation 13 in equation 17,

$$k_{eff} = \frac{H}{\frac{2(H - 2r)}{2k_p} + \frac{4r^2 S}{(k_p V_p + k_f V_f)}} = \frac{1}{\frac{1}{k_p} \left(1 - \left(\frac{6\phi}{\pi} \right)^{1/3} \right) + \frac{4r^2 H}{(k_p V_p + k_f V_f)}} \tag{18}$$

This can further be rearranged as,

$$k_{eff} = \frac{1}{\frac{1}{k} \left(1 - \left(\frac{6\phi}{\pi} \right)^{1/3} \right) + \frac{2}{\frac{1}{2r^2 H} (k_p V_p + k_f V_f)}} \tag{19}$$

Substituting the value of V_p from equation 11 in equation 19, the final expression for effective thermal conductivity is obtained as:

$$k_{eff} = \frac{1}{\frac{1}{k_p} \left(1 - \left(\frac{6\phi}{\pi} \right)^{\frac{1}{3}} \right) + \frac{2}{\left\{ k_p \left(\frac{4\pi}{3\phi} \right)^{\frac{1}{3}} + \pi \left(\frac{2\phi}{9\pi} \right)^{\frac{1}{3}} \left(k_f \frac{D_p}{D_f} - k_p \right) \right\}}} \quad (20)$$

The correlation given in the equation 19 can thus be used to estimate k_{eff} for particulate filled polymer composites with different filler content.

Experimental details

Material considered

Matrix Material. Epoxy LY 556 resin, chemically belonging to the ‘epoxide’ family is used as the matrix material. Its common name is Bisphenol-A-Diglycidyl-Ether. The low temperature curing epoxy resin (Araldite LY 556) and the corresponding hardener, Triethylene tetramine (TETA, HY 951) are mixed in a ratio of 10:1 by weight as recommended. The epoxy resin and the hardener used for the present work are supplied by Ciba Geigy Limited, India. Epoxy is chosen primarily because it happens to be the most commonly used polymer and also possesses low density (1.1 gm/cm^3). Also it possesses low thermal conductivity (0.363 W/m-K) which is the prime requirement for present work.

Filler material. Teak wood dust (TWD) is generated during the cutting of teak tree. Teak is found in the Himalayan subtropical region and this natural resource can be gainfully converted to value added products. The scientific name of teak is *Tectona grandis*. It is one of three species in the genus *Tectona*. *Tectona grandis* is native to India, Indonesia, Myanmar, northern Thailand, and northwestern Laos. Nilambur in Kerala, India is a major producer of teak of fine quality, holds the world's oldest teak plantation. Main organic constituents of teak

wood are: lignin, carbohydrates, hemi-cellulose, pentosans, ash, acetyl content and silica. Its dust particles appear as golden or medium brown, fibrous powder.

The teak wood dust of average particle size 100 μm is the reinforcing material in the present work. It is chosen mostly for its very low thermal conductivity (0.04 W/m K) and low density (0.8 gm/cc). Basically the wood dust is considered as a waste product. Moreover, it is renewable, eco-friendly and non-toxic. Also it has superb stability, good strength properties, easy workability and above all it has outstanding resistance to decay.

Composite fabrication

Hand lay-up is the simplest and oldest open molding method of the composite fabrication processes. All the epoxy based composite samples of various compositions are prepared by this route in the present investigation. Here low temperature curing epoxy resin (LY 556) and corresponding hardener (HY 951) are mixed in a ratio 10:1 by weight as recommended. Then teak wood dust of average particle size 100 μm is reinforced in the resin to prepare the composites in different proportions according to the experimental requirements. The uniformly mixed dough (epoxy filled with TWD) is then slowly decanted into the glass molds, coated beforehand with wax and a uniform thin film of silicone-releasing agent. Release agents facilitate removal of parts from molds. The composites are cast in these molds so as to get disc type specimens (diameter 20 mm, thickness 5 mm). Composites of different compositions with varying TWD content (Table 1) are fabricated. The castings are left to cure at room temperature for about 24 hours after which the glass molds are broken and samples are released.

Characterization

Physical Characterization

Density. The actual density (ρ_a) of composite is determined by the Archimedes principle using distilled water as the medium as per ASTM standard D 792. According to this principle, when

an object is immersed in a liquid the apparent loss in its weight is equal to the upthrust and this is equal to the weight of the liquid displaced. The density of the composite is obtained by using the following equation,

$$\rho_a = \frac{\rho_w W_a}{W_a - W_w} \quad (21)$$

Here ρ_a is the actual/measured density of composite, ρ_w is the density of distilled water, W_a is weight of the sample in air and W_w is weight of the sample in water. The theoretical density of composite materials (ρ_t) in terms of weight fraction can easily be obtained as per the following equations given by Agarwal and Broutman [24].

$$\rho_t = \frac{1}{\left(\frac{W_f}{\rho_p}\right) + \left(\frac{W_m}{\rho_m}\right)} \quad (22)$$

Where, W and ρ represent the weight fraction and density respectively. The suffix p and m stand for the filler and matrix materials respectively.

The void content in the composite is given by the following expression,

$$V_v = \frac{(\rho_t - \rho_a)}{\rho_t} \quad (23)$$

Scanning Electron Microscopy. The micro-structural features of the various particulate filled composite specimens are examined by Scanning Electron Microscope JEOL JSM-6480 LV. The specimens are mounted on stubs with silver paste. To improve the penetration of light and for better surface micrographs, a thin film of platinum is vacuum-evaporated onto the samples before the micro graphs are taken.

Mechanical Characterization

The tensile and compressive strength of the fabricated composites are performed as per ASTM E 1309 and ASTM D695 test standards respectively using a universal testing machine

Instron 1195. The cured rectangular shaped composite samples of required dimension (length 150 mm, width 20 mm and thickness 3 mm) are used for the tensile test and dimension (length 10 mm, diameter 10 mm) are used for the uni-axial compression test. The cross head speed of 10 mm/minute and 1mm/min is maintained for tensile and compression test respectively. The crosshead speed is maintained constant and is stopped when the specimen shows signs of failure or when densification is complete. The three point bend test is conducted on all the composite samples as per ASTM D7264 in the universal testing machine Instron 1195 for evaluating flexural strength. The dimension of each specimen is 60 mm × 10 mm × 3 mm. The experiment is conducted at a cross head speed of 10 mm/min. In all the above cases, the test is repeated three times on each composite type and the mean value is reported.

Thermal Characterization

Thermal Conductivity. Unitherm™ Model 2022 is used to measure thermal conductivity of a variety of materials. These include polymers, ceramics, composites, glasses, rubbers, some metals and other materials of low to medium thermal conductivity. The tests are done in accordance with ASTM E-1530 standard. Circular sample of 20 mm diameter and 3 mm of thickness with flat surface on both side are used for the measurement. This instrument works on the principle of guarded heat flow meter test method.

Glass transition temperature and coefficient of thermal expansion. Glass transition temperature and coefficient of thermal expansion of the composites are measured with a Perkin Elmer DSC-7 Thermal Mechanical Analyzer (TMA) in expansion mode. At first, the TMA sample stage is purged with nitrogen gas. The sample length is set between 6-8 mm and the width as well as thickness is about 2-3 mm. During the TMA measurement, the specimen is heated from 30 to 150°C at a heating rate of 5°C/min. For each measurement, two heating scans are used. The first heating scan is used to eliminate any possible internal stress and moisture in the sample which is likely to be generated during the curing and sample

preparation processes. The second heating scan is used to determine the T_g and CTE of the material.

Results and discussion

Physical Characterization

Density and volume fraction of voids. The theoretical and measured densities along with the corresponding volume fraction of voids in the epoxy-TWD composites are presented in Table 1. It may be noted that the composite density values calculated theoretically from weight fractions using equation 21 are not equal to the experimentally measured values. This difference is a measure of voids and pores present in the composites. It is found that with the increase in TWD content in epoxy resin from 0 to 30 vol%, there is a drop in density of the composite by about 9.2 % although there is a simultaneous increase in the void fraction or porosity to 1.815% (Table 2).

Density of a composite depends on the relative proportion of matrix and reinforcing materials and this is one of the most important factors determining the properties of the composites. The void content is the cause for the difference between the values of true density and the theoretically calculated ones. The voids significantly affect some of the mechanical properties and even the performance of the composites in the workplace. The knowledge of void content is desirable for better estimation of quality of the composites.

Micro-structural characterization. It is well known that the properties of the composites are strongly dependent on the interaction between the filler and the matrix. In order to evaluate this interaction, the microstructure of the composites including the dispersion of the micro-sized teak wood dust particles in the epoxy matrix is observed under scanning electron microscope. Figure 4 (a) shows a typical SEM image of TWD filled epoxy composite. A single TWD particle (within the matrix body) at a higher magnification is shown in Figure 4

(b). These micrographs confirm the near-spherical shape of the filler particles i.e. TWD used as reinforcement in the polymer resin.

Mechanical Characteristics

Tensile, compressive and flexural strength of the composite specimens are evaluated and the test results for all these composites are presented in Table 3. It is noticed that with addition of TWD particles, tensile strength of the composite decreases and this decrement is a function of the filler content. It is noted that while the tensile strength of neat epoxy is 65 MPa, it decreases by about 9.4 % to 58.9 MPa with the incorporation of 30 vol % of TWD in it. It is again noticed that with addition of TWD particles, compressive strength of the composite also decreases and this decrement is a function of the filler content. While the compressive strength of neat epoxy is 90MPa, it drops by about 8.5 % to 82.30 MPa for maximum filler loading. Composite materials used in structures are prone to fail in bending and therefore development of new composites with improved flexural characteristics is essential. It is further noticed that flexural strength of the composite decreases with addition of TWD particles. While the flexural strength of neat epoxy is 58MPa, it decreases by about 5.8 % to 54.60 MPa for maximum filler loading.

There can be two reasons for the reduction of various strength properties as the teak wood dust content increases. The first reason may be due to the fact that the chemical reaction at the interface between the filler particle and the matrix is too weak to transfer the tensile stress; the second reason may be due to the rise of the stress concentration at the sharp corner of some of the irregularly shaped particulates embedded in the epoxy matrix.

Thermal Characteristics

Effective Thermal Conductivity (k_{eff})

Numerical Method. Figure 5 clearly illustrate the heat flow direction and the boundary conditions for the particulate-polymer composite body considered for the analysis of this conduction problem. The temperature at the nodes along the surface ABCD is T_1 ($=100^0\text{C}$), the ambient convective heat transfer coefficient is assumed to be $2.5 \text{ W/m}^2\text{-K}$ and the analysis of the problem is done at a room temperature of 27^0C . The other surfaces parallel to the direction of the heat flow are assumed to be adiabatic. The temperatures at the inside domain and on the other boundaries are not apprehended. The unknown temperatures at the interior nodes and on the other boundaries are obtained with the help of ANSYS.

A few presumptions involved in this analysis are:

- a) The composites are macroscopically homogeneous;
- b) Locally both the matrix and filler are homogeneous and isotropic;
- c) The filler-matrix interfacial thermal contact resistance is negligible;
- d) The composite body is free from voids;
- e) The fillers are assumed to be uniformly distributed in the matrix in a square periodic array.

Thermal conductivities of the TWD-epoxy composites are numerically estimated by using the sphere-in-cube model. Some typical 3-D physical models showing the arrangement of TWD micro spheres within the cube shaped composites with filler concentrations of 6.5 vol % and 26.8 vol % are illustrated in Figures 6. Similarly, the temperature profiles obtained from FEM analysis for composites with TWD concentrations of 1.41, 3.4, 6.5, 11.3, 17.95 and 26.8 vol % are presented in Figures 7 (a) to (f) respectively.

From the temperature profiles, the value of effective thermal conductivity of each composite sample is estimated on the basis of one dimensional heat conduction equation. As expected,

the values of k_{eff} come out to be different for composites with different TWD content. These values obtained from FEM simulation for composites with different filler concentrations ranging from 1.41 to 26.8 vol% are presented in Table 4. It also presents the corresponding k_{eff} values calculated by using various existing theoretical correlations such as Maxwell's and Lewis and Nielsen's model for composites having same filler concentrations.

The measured values of effective thermal conductivity of epoxy-TWD composites for different filler content are also presented in Table 4 along with the values obtained from the proposed model. It is seen from Table 4 that the values of effective thermal conductivity of composites obtained from different theoretical correlations are in good agreement with the values obtained from FEM model. With the increase in TWD content, a monotonic decrement in the value of k_{eff} is noticed irrespective of the method or model employed for the purpose. But large discrepancies are noted between these values and the experimentally measured ones.

It is observed that the experimentally measured k_{eff} of the composites are significantly lower than all the theoretically predicted values irrespective of the filler concentration. This can partly be attributed to the presence of voids and pores within the composite body but the more possible reason is the thermal resistance at the filler-matrix interface. Polymers usually have hydrophobic surfaces therefore fillers with polar surfaces such as TWD would lead to poor interfacial bonding to the polymer further leading to high thermal contact resistance at the interfacial region. It is believed that the thermal contact resistance at the interface (R_{int}) plays a dominant role in significantly hindering the heat conduction capability of the composite system.

In the present work the proposed correlation (equation 20) does not consider the effect of thermal contact resistance at the interfacial region and hence overestimates the value of k_{eff} for composites with different filler concentrations. It is further noticed that incidentally the

difference between the predicted and measured values of k_{eff} for any particular filler volume fraction happens to be proportional to the ratio of total filler-matrix interfacial area (A_{int}) to the volume of the composite (V). Therefore a new term $\psi * \left(\frac{A_{int}}{V} \right)$ is introduced to the correlation to improve the accuracy of theoretical prediction. Here, ψ is the proportionality constant and the new term in the modified correlation (Equation 24) takes into account the reduction in effective thermal conductivity of the composite due to interfacial thermal contact resistance.

$$k_{eff} = \frac{1}{\frac{1}{k_p} \left(1 - \left(\frac{6\phi}{\pi} \right)^{\frac{1}{3}} \right) + \frac{2}{\left\{ k_p \left(\frac{4\pi}{3\phi} \right)^{\frac{1}{3}} + \pi \left(\frac{2\phi}{9\pi} \right)^{\frac{1}{3}} \left(k_f \frac{D_p}{D_f} - k_p \right) \right\}}} - \psi \left(\frac{A_{int}}{V} \right) \quad (24)$$

Using the above correlation, the effective thermal conductivities of the composites under study are estimated for different filler concentrations. In order to validate the correlation and to attain a close approximation with the experimental results a suitable value for ψ as 11×10^{-6} is chosen. Table 5 reflects the variation in ratio of total filler-matrix interfacial area (A_{int}) to the volume of the composite (V) for different filler content. Figure 8 presents the values of k_{eff} obtained from different theoretical models. These values are compared with the experimentally measured values of k_{eff} for epoxy-TWD composites of different compositions.

In order to further validate the proposed correlation, the thermal conductivities of SET 2 composites are measured experimentally using the same Unitherm™ 2022 conductivity tester. The measured values are compared with the theoretically estimated values (Equation 24) and the percentage error associated with each predicted values is presented in Table 6.

It is evident from Table 6 that the predicted values of effective thermal conductivities based on the proposed correlation with $\psi = 11 \times 10^{-6}$ are in close approximation with the measured values for epoxy-TWD composites with a wider range of filler concentrations (0-30 vol %). It

may thus be stated that Equation 24 can be used as a predictive equation for other particulate-polymer composite systems by suitably tuning the value of ψ .

It is seen that incorporation of micro-sized TWD particles into epoxy appreciably enhances its heat insulation capability. Experimental results suggest that by adding 30 vol % of TWD, the thermal conductivity of epoxy can be reduced by about 84% (from 0.363 to 0.058 W/mK). It also becomes clear that this reduction is attributed not only to the relatively lower conductivity of TWD but also to the thermal contact resistance offered by the interfacial region of filler-matrix combination.

Glass Transition Temperature (T_g). Figure 9 presents the variation of T_g with TWD content. It is observed that the T_g of neat epoxy is about 98°C and it gradually increases to 109°C with an increase in TWD content. A maximum increase of 11°C in T_g is obtained as the TWD content in the composite is increased from 0 to 30 vol %. The effect of filler addition in polymer composites on glass transition behavior has been investigated in the past for different particulate filled polymer composites [25, 26] and it has been seen that addition of micro-sized fillers usually improves the T_g of composites [26]. In the present study, a shift of T_g towards a higher value can be attributed to the filler-matrix interaction, which might be restricting the mobility of the epoxy chain.

Coefficient of Thermal Expansion (CTE). Figure 10 shows that the CTE of the composite reduces with increase in TWD content. The CTE of neat epoxy is about 66 ppm/⁰C and it gradually reduces to 63.5 ppm/⁰C with increase in filler content. A maximum decrease of about 3.79 % in CTE is obtained with 30 vol % of TWD in the composite. The constraint of mobility of the epoxy chain due to the interaction of TWD and epoxy are responsible for the reduction of CTE. Hence, the thermal stability of epoxy improves with addition of TWD particles.

Conclusions

This analytical and experimental investigation on epoxy composites filled with TWD micro-particles led to the following specific conclusions:

1. Fabrication of epoxy-TWD composites by hand layup technique is possible.
2. Incorporation of TWD particles modifies the tensile, compressive and flexural strengths of the composites. It is noticed that with addition of TWD particles, the strength of epoxy is marginally decreased and this decrement is a function of the filler content.
3. A theoretical correlation to estimate the effective thermal conductivity of particulate filled composites is proposed based on the one dimensional heat conduction model. It is seen that this serves as a good empirical model for spherical inclusions and that the proposed correlation can therefore very well be used to estimate the effective thermal conductivity for composites.
4. It is propitious that the incorporation of TWD particles results in significant improvement in the thermal insulation capability of epoxy resin. It is found that with increase in the TWD content from 0 to 30 vol %, the value of effective thermal conductivity (k_{eff}) drops by about 84% for epoxy.
5. It is observed that the T_g of epoxy (98°C) gradually increases to 109°C as the TWD content increases from 0 to 30 vol %. It is further seen that with the incorporation of TWD, the coefficient of thermal expansion (CTE) of epoxy is substantially lowered.
6. With light weight, lowered thermal expansion coefficient and improved insulation capability, the teak wood dust filled epoxy composites can be used for applications such as insulation boards, food containers, thermos flasks, refrigeration industry, building materials, interiors of air crafts and automobiles etc.

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Table 1. Epoxy composites filled with teak wood dust

S.No.	Composition	
	SET I	SET II
1	Epoxy + 1.4 vol % TWD	Epoxy + 5 vol % TWD
2	Epoxy + 3.4 vol % TWD	Epoxy + 10 vol % TWD
3	Epoxy + 6.8 vol % TWD	Epoxy + 15 vol % TWD
4	Epoxy + 11.3 vol % TWD	Epoxy + 20 vol % TWD
5	Epoxy + 17.95 vol % TWD	Epoxy + 25 vol % TWD
6	Epoxy + 26.8 vol % TWD	Epoxy + 30 vol % TWD

Table 2. Measured and Theoretical densities of the composites

Composition	Measured Density (gm/cc)	Theoretical Density (gm/cc)	Volume Fraction of Voids (%)
Epoxy + 5 vol % TWD	1.071	1.085	1.307
Epoxy + 10 vol % TWD	1.060	1.070	0.943
Epoxy + 15 vol % TWD	1.042	1.055	1.248
Epoxy + 20 vol % TWD	1.024	1.039	1.465
Epoxy + 25 vol % TWD	1.008	1.022	1.389
Epoxy + 30 vol % TWD	0.992	1.010	1.815

Table 3. Mechanical properties of the fabricated composites

Composition	Tensile Strength (MPa)	Flexural Strength (MPa)	Compressive Strength (MPa)
Neat epoxy	65.00	58.00	90.00
Epoxy + 5 vol % TWD	64.50	57.70	88.20
Epoxy + 10 vol % TWD	63.70	57.20	86.80
Epoxy + 15 vol % TWD	62.80	56.60	85.50
Epoxy + 20 vol % TWD	61.70	55.80	84.60
Epoxy + 25 vol % TWD	60.30	55.30	83.40
Epoxy + 30 vol % TWD	58.90	54.60	82.30

Table 4. Effective thermal conductivities of composites: Theoretical, numerical and measured values

Composition	Effective thermal conductivity(W/mK)				
	Maxwell's model	Lewis and Nielsen model	FEM model	Proposed model	Measured values
Epoxy + 1.41 vol% TWD	0.357	0.356	0.357	0.359	0.333
Epoxy + 3.4 vol% TWD	0.347	0.346	0.348	0.352	0.320
Epoxy + 6.5 vol% TWD	0.334	0.331	0.335	0.342	0.290
Epoxy + 11.3 vol% TWD	0.313	0.306	0.316	0.326	0.240
Epoxy + 17.95 vol% TWD	0.286	0.273	0.296	0.304	0.187
Epoxy + 26.8 vol% TWD	0.252	0.232	0.277	0.276	0.092

Table 5. Variation in ratio of total filler-matrix interfacial area to the volume of the composite

Composition	A_{int}/V (m^{-1})
Epoxy + 1.41 vol % TWD	846
Epoxy + 3.4 vol % TWD	2040
Epoxy + 6.5 vol % TWD	3900
Epoxy + 11.3 vol % TWD	6780
Epoxy + 17.95 vol % TWD	10700
Epoxy + 26.8 vol % TWD	16080

Table 6. Comparison of theoretical and experimental k_{eff} values of Epoxy-TWD composite

Composition	Effective Thermal Conductivity (W/m-K)		Error Percentage (%)
	Measured Value	Proposed Correlation (Modified)	
Epoxy + 5 vol % TWD	0.309	0.314	1.618
Epoxy + 10 vol % TWD	0.253	0.264	4.348
Epoxy + 15 vol % TWD	0.202	0.214	5.941
Epoxy + 20 vol % TWD	0.148	0.165	11.486
Epoxy + 15 vol % TWD	0.098	0.116	18.367
Epoxy + 30 vol % TWD	0.058	0.066	19.846



Figure 1. (a) 3-D view of particulate filled composite cube (b) 3-D view of element under study

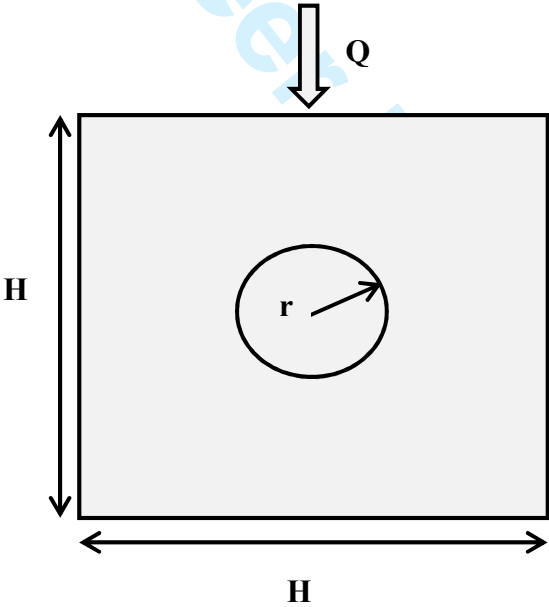


Figure 2. Heat transfer model for particulate-filled polymer composite

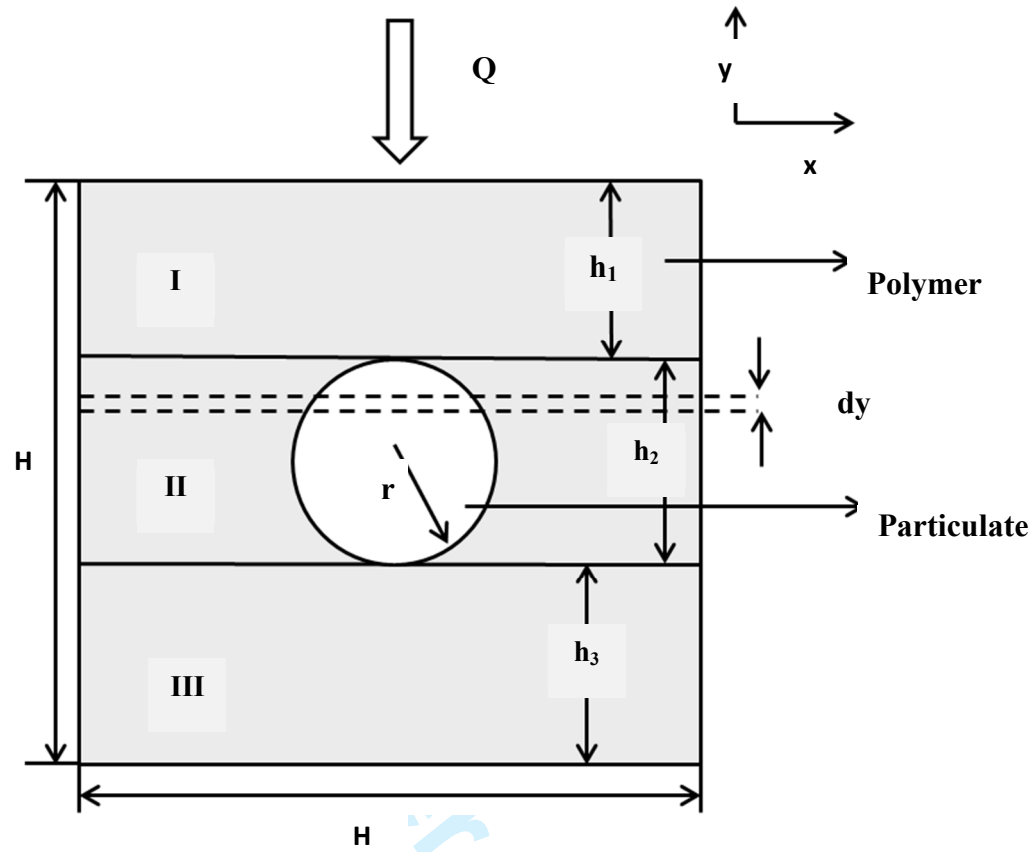
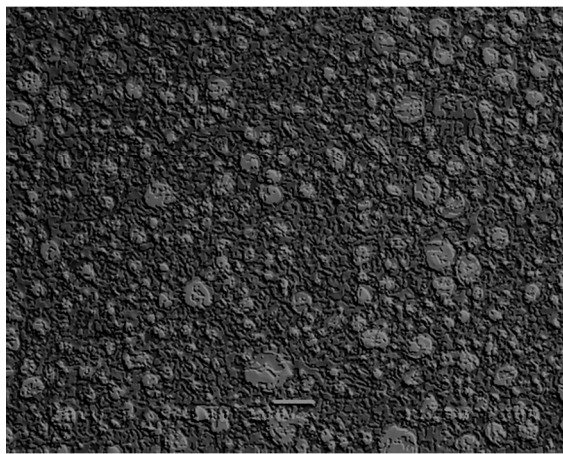
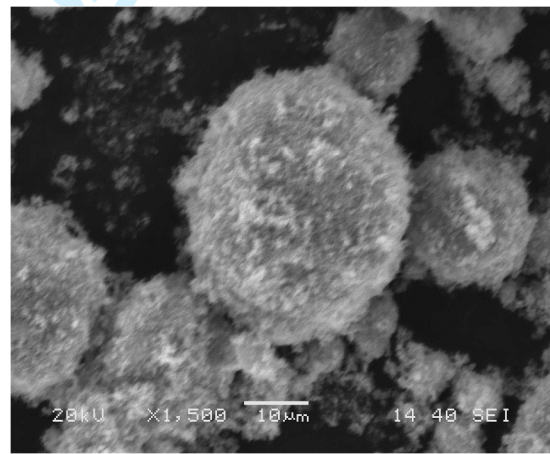


Figure 3. Series model of heat transfer in particulate-filled composite



(a)



(b)

Figure 4. SEM images of epoxy composites filled with micro-sized TWD particles (a) lower magnification (b) Higher magnification

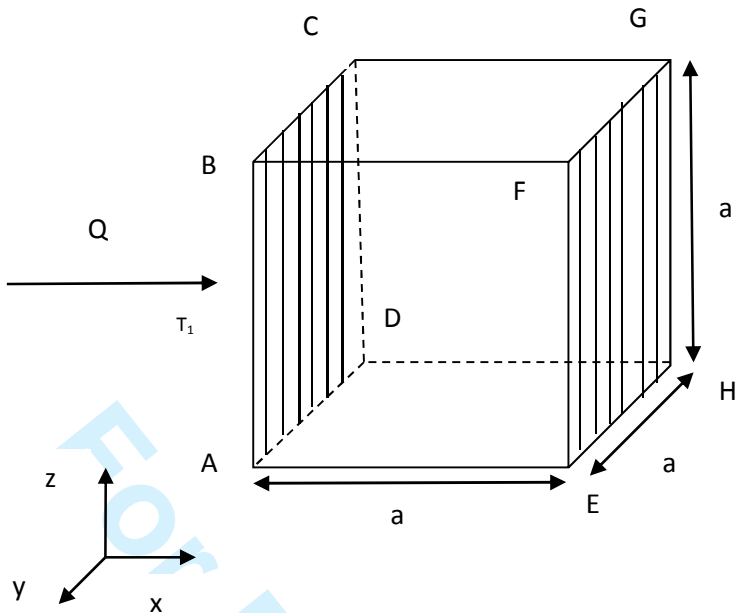


Figure 5. The heat flow direction and boundary conditions for the particulate - polymer composite

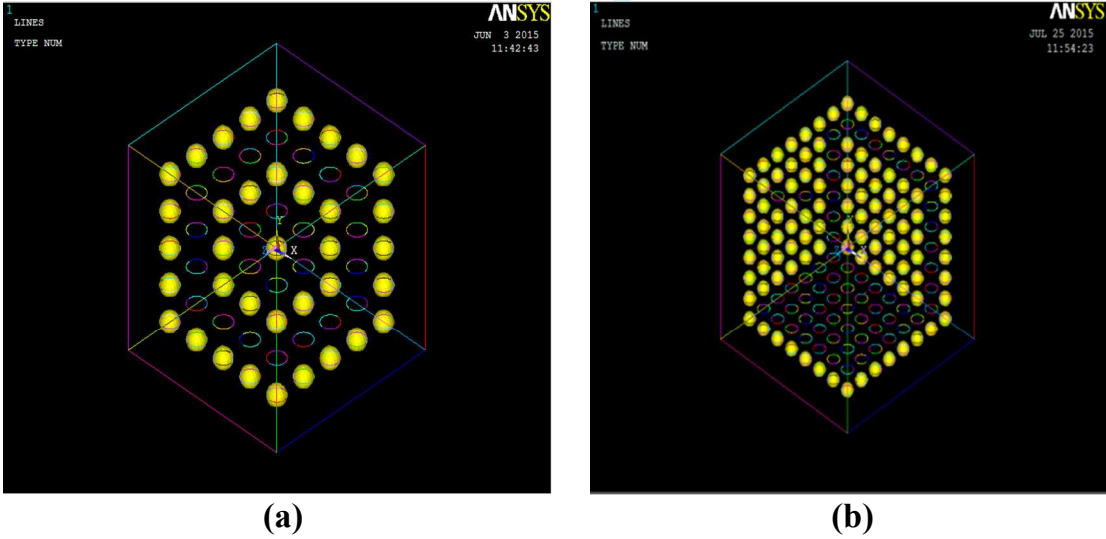
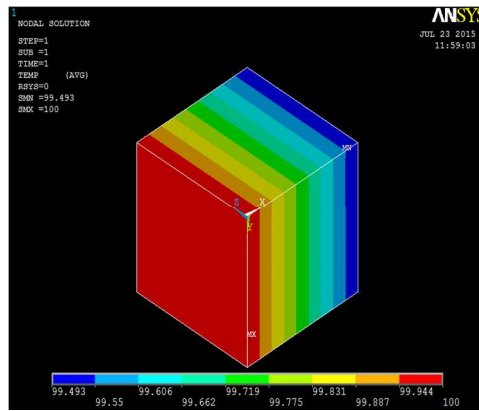
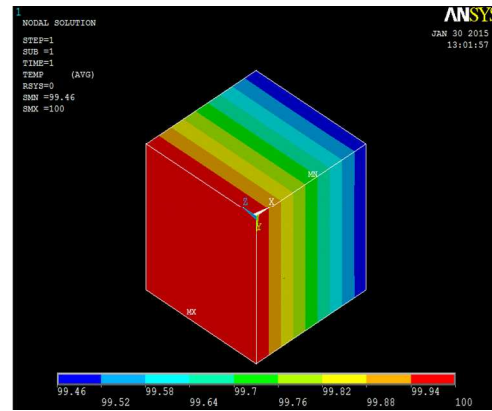


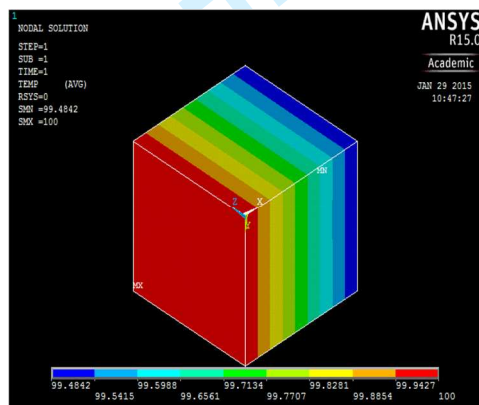
Figure 6. Typical 3-D sphere-in-cube models for composites with different filler concentrations
(a) 6.5 vol % (b) 26.8 vol %



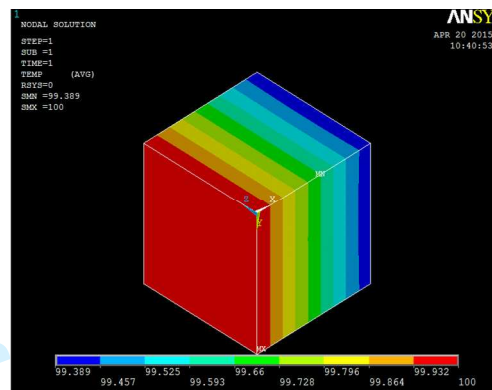
(a)



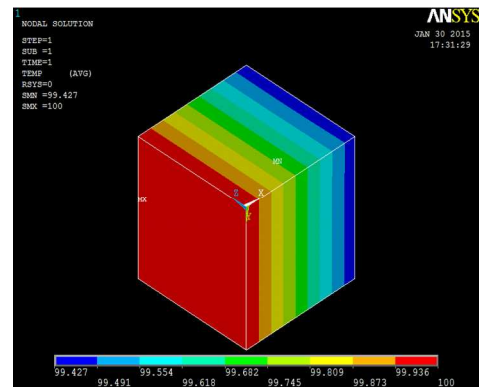
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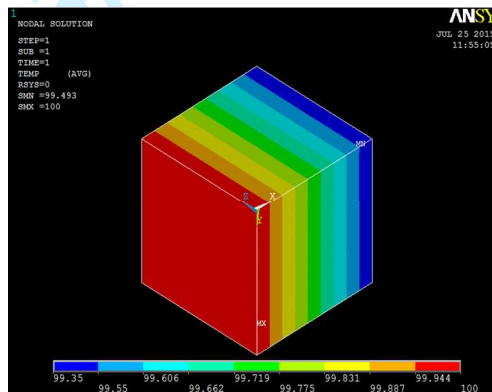
(c)



(d)



(e)



(f)

Figure 7. Temperature profiles obtained for epoxy filled with (a) 1.41 vol % TWD (b) 3.4 vol % TWD (c) 6.5 vol % TWD (d) 11.3 vol % TWD (e) 17.95 vol % TWD (f) 26.8 vol % TWD

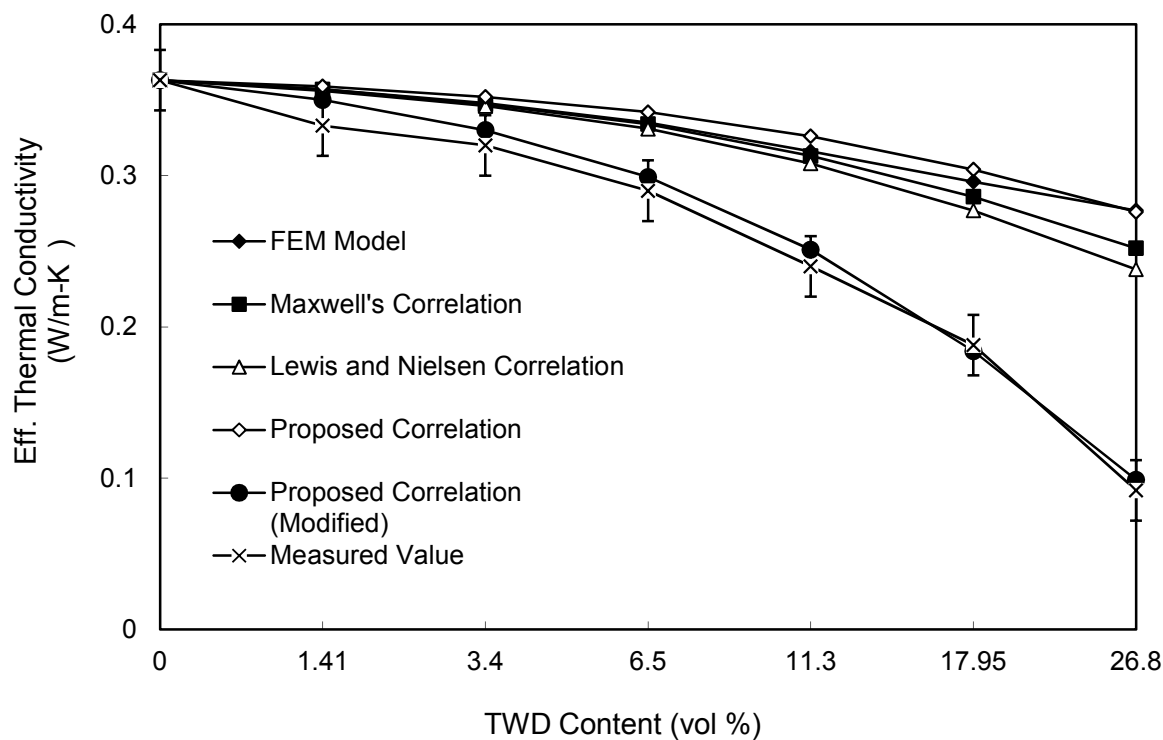


Figure 8. Comparison of k_{eff} values obtained from various methods

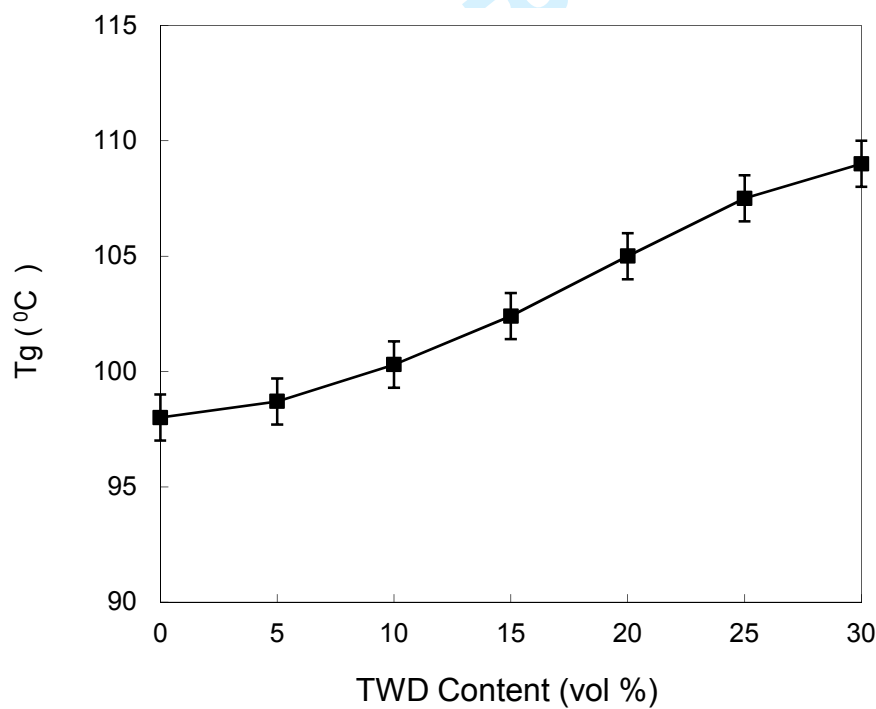


Figure 9. Variation of T_g of epoxy-TWD composites with filler content

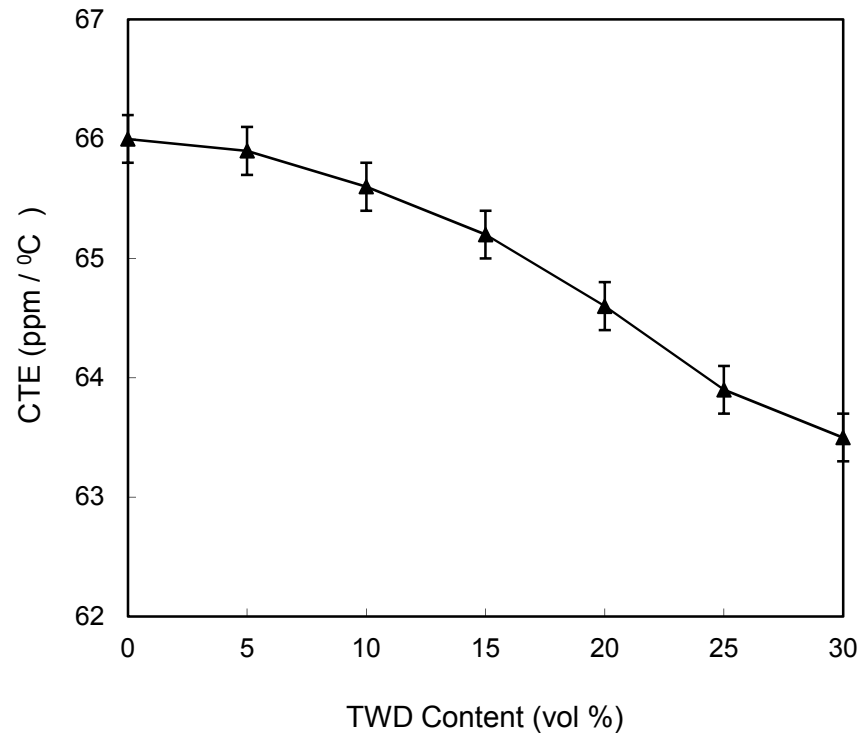


Figure 10. Variation of CTE of epoxy-TWD composites with filler content